ACID RAIN: IMPLICATIONS FOR FOSSIL R&D

HEARINGS

BEFORE THE

SUBCOMMITTEE ON
ENERGY DEVELOPMENT AND APPLICATIONS

AND THE

SUBCOMMITTEE ON NATURAL RESOURCES,
AGRICULTURE RESEARCH AND ENVIRONMENT

OF THE

COMMITTEE ON
SCIENCE AND TECHNOLOGY
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ACID RAIN: IMPLICATIONS FOR FOSSIL R&D

TUESDAY, SEPTEMBER 13, 1983

HOUSE OF REPRESENTATIVES, COMMITTEE ON SCIENCE AND TECHNOLOGY, SUBCOMMITTEE ON ENERGY DEVELOPMENT AND APPLICATIONS; AND SUBCOMMITTEE ON NATURAL RESOURCES, AGRICULTURE RESEARCH AND ENVIRONMENT,

Washington, D.C.

The subcommittees met, pursuant to call, at 9:30 a.m., in room 2318, Rayburn House Office Building, Hon. Don Fuqua (chairman of the Subcommittee on Energy Development and Applications) presiding.

Mr. FUQUA. The meeting will come to order.

It's a pleasure to welcome our witnesses and guests to our first day of hearings on acid rain. The issue of acid rain is emerging as one of the most important environmental issues facing the 1980's.

There is a strong consensus in the scientific community that the vast majority of the undesirable acidic end products of wet and dry deposition—sulfates and nitrates—is derived from the combustion of fossil fuels, which release our sulfur dioxide and nitrogen oxides into the atmosphere.

On the other hand, there appears to be a great deal of disagreement and uncertainty as to the details of the causes, effects, and control of acid deposition.

At a time when there is mounting political pressure to mitigate the impacts of acid deposition, there is, in particular, little agreement on which combination of developed or developing technologies might be used to control the sulfur and nitrogen emissions which contribute to the acid rain problem.

We hope, through the course of these hearings, to address three principal questions:

First, what is the state of knowledge of the causes, effects, and control of acid rain?

Second, what is the status of current research programs and plans in these areas? And,

Third, how may developed and developing control technologies be used in a cost-effective manner to effect reductions which may be mandated in sulfur and nitrogen emissions?

Today, we are looking forward to hearing testimony from our distinguished panel of witnesses about the current state of knowledge, as well as the status and current plans of the national acid precipitation assessment program.

Our first witness will be Dr. J. Christopher Bernabo——

Dr. BERNABO. Bernabo.

Mr. FUQUA. Bernabo, Director of Research for the Interagency Task Force; followed by Dr. Jack Calvert, of the National Center for Atmospheric Research and Chairman of the National Council's Committee from Boulder; and Dr. William C. Ackermann of the Department of Civil Engineering at the University of Illinois, Urbana-Champaign campus.

And before we begin, we will—without objection, grant permission for television and recording equipment.

Dr. Bernabo.

STATEMENT OF DR. J. CHRISTOPHER BERNABO, EXECUTIVE DIRECTOR OF THE NATIONAL ACID PRECIPITATION ASSESSMENT PROGRAM

Dr. BERNABO. Thank you, Mr. Chairman.

I welcome the opportunity to be here today to discuss with you the national acid precipitation assessment program and I would like to request that my written statement be entered in full in the record and that I be allowed to make a more spontaneous presentation using some slides.

Mr. FUQUA. That will be granted, and if you wish to summarize, certainly feel free.

Dr. BERNABO. Yes. Thank you.

The national acid precipitation assessment program is the statutorily mandated effort which pulls together the activities of all the Federal agencies in acid rain research and attempts to integrate them into a comprehensive program examining the causes, effects, and possible solutions to acid rain.

Now, in this program, we have involved literally hundreds of scientists, not only in Federal agencies, but also in universities, the State agencies, and private-sector groups, and the program involves not only research, but also monitoring and assessment activities, looking at the acid deposition problem.

Now, if I might switch slides here—I don't know—if somebody could help me with the lights——

Mr. FUQUA. Before you get started, I inadvertently omitted—Congressman Winn had asked permission to insert a statement following one I have, and without objection, that will be inserted there.

[The opening statement of Mr. Winn and Mr. Sensenbrenner follow:]

OPENING STATEMENT OF THE
HON. LARRY WINN, JR. (R-KAN.)

HEARINGS ON ACID RAIN:
IMPLICATIONS FOR FOSSIL ENERGY R&D

TUESDAY, SEPTEMBER 13, 1983

THANK YOU, MR. CHAIRMAN, FOR THIS OPPORTUNITY TO MAKE A BRIEF STATEMENT. YOUR HEARINGS BOTH TODAY AND NEXT WEEK ARE CERTAINLY FITTING AND TIMELY.

WE ARE ALL WELL AWARE OF THE GROWING DISCUSSION ON ACID RAIN; QUESTIONS ABOUT WHAT CAUSES IT, WHAT THE EFFECTS ARE, AND HOW RAPIDLY AND HOW COMPLETELY WE SHOULD CONTROL IT. THAT IS WHY IT IS EXTREMELY IMPORTANT THAT RECENT TECHNICAL REPORTS ON THE SUBJECT BE REVIEWED, AND THAT WE DETERMINE ON A COST-EFFECTIVE MANNER THE EXTENT TO WHICH ACID RAIN CAN BE REDUCED.

ACTUALLY, WE WILL BE ADDRESSING THE LARGER ASPECT, WHICH IS TERMED "ACID DEPOSITION". THIS INCLUDES NOT ONLY ACID RAIN, BUT ALSO THE SOLID PARTICULATE WHICH CONTAINS ACID-FORMING CHEMICALS. UNFORTUNATELY, THIS COMPLICATES THE MATTER. BUT, THERE ARE GREAT LEGITIMATE ENVIRONMENTAL CONCERNS WITH LAND

AND WATER, AND WE ARE VERY INTERESTED IN ANY MATERIAL THAT FALLS FROM THE ATMOSPHERE.

A FURTHER COMPLICATION IS THAT WE HAVE BEEN MEASURING EMISSIONS TO, AND PRECIPITATION FROM, THE ATMOSPHERE FOR A RELATIVELY SHORT PERIOD OF TIME. THEREFORE, WE HAVE A MEAGER AMOUNT OF INFORMATION WITH WHICH TO WORK. THE RESEARCH FUNDS CURRENTLY BEING PROVIDED BY THIS COMMITTEE WILL ASSURE THE ADDITIONAL DATA SO BADLY NEEDED TO PROVIDE FOR THE NECESSARY TECHNICAL JUDGMENTS WHICH MUST BE MADE.

MR. CHAIRMAN, WE ARE ALL INTERESTED IN THE IMPROVEMENT OF THE WORLD IN WHICH WE LIVE. WE WOULD LIKE TO CORRECT ALL DEFICIENCIES IMMEDIATELY. AT THE SAME TIME, THERE IS A COST THAT GOES WITH THESE CORRECTIVE ACTIONS. WE HAVE ALL SEEN THE WASTE OF MANPOWER AND MONEY THROUGH MISDIRECTION IN THE PAST. IT IS CERTAINLY HOPED THAT WE CAN GO FORWARD WITH ACID DEPOSITION RELIEF AND THAT OUR DIRECTION MAY BE MADE EFFECTIVE THROUGH THE USE OF DEFINITIVE DATA.

THANK YOU, MR. CHAIRMAN. I LOOK FORWARD TO HEARING FROM OUR WITNESSES.

OPENING STATEMENT OF THE
HON. F. JAMES SENSENBRENNER, JR. (R-WIS.)

HEARINGS ON ACID RAIN:
IMPLICATIONS FOR FOSSIL ENERGY R&D

TUESDAY, SEPTEMBER 13, 1983

THANK YOU, MR. CHAIRMAN, FOR THIS OPPORTUNITY TO EXTEND A WELCOME TO OUR DISTINGUISHED WITNESSES.

WE LOOK FORWARD TO THEIR TESTIMONY ON THE STUDIES THAT THEIR RESPECTIVE ORGANIZATIONS HAVE COMPLETED ON THE SUBJECT OF ACID DEPOSITION IN THE UNITED STATES.

ACID DEPOSITION IS A SERIOUS ENVIRONMENTAL PROBLEM WHOSE EFFECTS ARE WELL ESTABLISHED, BUT WHOSE CAUSES HAVE BEEN SUBJECT TO DISPUTE. WHILE THE FOCUS OF ATTENTION REGARDING THE EFFECTS OF ACID DEPOSITION HAVE CENTERED ON CANADA AND THE NORTHEAST IN THIS COUNTRY, WE IN WISCONSIN HAVE BEEN CONCERNED ABOUT ITS IMPACT UPON OUR LAKES AND RIVERS. IN 1980, THE JOINT ACID DEPOSITION TECHNICAL REVIEW COMMITTEE WAS FORMED, CONSISTING OF REPRESENTATIVES FROM THE STATE'S DEPARTMENT OF NATURAL RESOURCES, THE PUBLIC SERVICE COMMISSION, AND THE WISCONSIN UTILITIES, TO CONDUCT RESEARCH ON THIS SUBJECT IN THE STATE. THIS PROGRAM WAS DESCRIBED BY

THE U.S. INTERAGENCY TASK FORCE ON ACID PRECIPITATION AS "AN EXEMPLARY STATE PROGRAM (WHICH) REFLECTS AN INTEGRATED EFFORT BY STATE AND PRIVATE SECTOR GROUPS." THE WISCONSIN PROGRAM HAS RECENTLY ISSUED ITS FIRST REPORT ON THIS SUBJECT. I WOULD ASK, MR. CHAIRMAN, THAT THE SUMMARY OF THE FIRST ANNUAL REPORT ON THE WISCONSIN ACID DEPOSITION RESEARCH PROGRAM BE INCLUDED IN THE RECORD AT THE CONCLUSION OF MY STATEMENT AND THAT THE FULL REPORT BE AVAILABLE FOR COMMITTEE CONSIDERATION OF THIS SUBJECT.

THANK YOU, MR. CHAIRMAN.

Mr. FUQUA. I apologize for interrupting.

Dr. BERNABO. Yes.

OK, as I think you are all well aware, acid rain is a highly controversial topic. It is controversial politically and it is controversial scientifically.

The point I would like to make here, though, is the situation is such that we have a lot of high-toned rhetoric and that, in fact, the politics and the demands for answers from policymakers are far exceeding at this point what the scientists can really provide—not that they can't provide anything; it is just that science is somewhat slower at moving forward than is the politics.

This leaves a situation where a number of advocates on different sides of the issue throw a lot of things into the discussion, but I think what there has been a relative lack of is real objective analysis of what this state of the information is. I think it is very important that you have today here one of our distinguished people who are not involved in the policy process, but who can give you some insights to where the science stands.

[See slide 1, p. 20.]

In recognition of this need for the scientific basis for formulating policy, the Congress, of course, enacted the Acid Precipitation Act in 1980. This legislation established the interagency task force responsible for managing this effort. There are 12 Federal agencies involved and the effort is chaired by a troika, that is, including the Environmental Protection Agency, the National Oceanic and Atmospheric Administration, the Department of Agriculture.

This management board, or board of directors, if you will, includes people typically at the assistant administrator or assistant secretary level in these agencies responsible for the research.

[See slide 2, p. 21.]

We have then created—and you see down at the bottom there a series of boxes—a set of task groups. These are actual scientists, project managers, working on a wide range of different aspects to acid rain, ranging from the sources, both natural and manmade, at-

mospheric processes, the various effects of aquatic, terrestrial, and materials. We are examining the possible health effects. For instance, the drinking water and food chains in the aquatic and terrestrial areas.

We are also looking at the possible control technologies applicable to alleviating the acid rain problem, as well as a group which does assessment and pulls together the scientific information in a form that can be used by policymakers and we also have an international group, because as you are aware, there is a lot of international ramifications and much research going on in other nations on acid rain.

[See slide 3, p. 22.]

What I am going to try and do then is go through these different research areas, give you a very brief idea of what the consensus in the scientific community is on the status of the science, and also what research we're doing to try and improve the information base.

[See slide 4, p. 23.]

This task force responsible for the national program has a number of responsibilities. The first of those is to develop a 10-year national plan. That was submitted to Congress back in 1982 to provide a broad framework for this long-term effort.

We are doing subsequent planning for that in a much more detailed basis and leaving our planning operation very flexible. As the Congress, the administration and others move forward with developing national policy, we feel the research should be responsive to supporting those policy options and being able to provide the information most relevant to them.

The task force also is responsible, then, in implementing this program and that involves, obviously, coordinating diverse efforts of the Federal agencies. We have a rather unique interagency budget process where all the agencies get together into a single budget process to avoid duplication and to make sure that major gaps in the program are avoided.

We maintain an inventory of all the federally sponsored research. We are also now putting onto our inventory State-sponsored research, particularly the State of California, and State of Wisconsin have major programs and other—a number of other States have programs, and the private sector, through the efforts of the Electric Power Research Institute are also inventorying those efforts, so we will have one data base for both managers and scientists alike all of the ongoing research in acid rain.

And we are encouraging and working with these various non-Federal groups in introducing the program. Again, there are major efforts, such as the ones by EPRI in the States which I have mentioned, which are important and which must be integrated into and interfaced by the Federal effort.

[See slide 5, p. 24.]

Beyond that, although we have a 10-year authorization for the program, we are charged in legislation with annually reporting to the President and Congress—and you have before you, and I have been asked to, in fact, report today on our annual report for 1982.

And 1982 was the first year in which we had budget authority and which was the first year of research. I am reporting to you on

the first year of research within this 10-year program and we will, in fact, be doing this reporting to Congress and the President on all the years moving out from here.

Finally, we are responsible for insuring that our research program is coordinated with the different international efforts. I think it is not commonly know, but the Canadians have a major acid rain research program of about $20 million a year, roughly equal to our own, and we have very excellent relations with them.

We have met with them once a year during the last 2 years, how to coordinate, to sponsor joint projects and to mutually work on the issue of the science of acid rain.

Now, again, as you are aware, there are many different aspects to the understanding of acid deposition and our program is charged specifically in the legislation with comprehensively examining everything from the sources to possible effects and mitigation aspects, so I am going to quickly again go through these different parts, if you will, of the acid rain cycle and discuss with you what information we have available and what research is underway.

Now, the area of natural sources—there is no question that there are major sources of natural material which contribute to acidity. On a global basis, it is believed that roughly half of the sulfur in the atmosphere is from natural sources. A single major eruption, such as the one of El Chichan, injected into the atmosphere as much sulfur as an entire year of man's activities in burning fossil fuels. However, these—the importance of these sources varies very much geographically, and in the areas of prime concern—that is, in northern Europe and in northeastern America, there is little doubt that the majority of the acidic substances are from man-made sources. In fact, it is estimated—and this is a map showing the emissions density—that sulfur nitrogen oxides contribute greater than 90 percent of the acidity occuring in northeastern North America.

So again, it is a question of where you are relative to the sources and it is clear in these industrialized areas and downwind of them, that man-made sources are the primary factors.

We are examining inventories of these sources and other substances and trying to improve our ability to project changes in them in the future based on control scenarios, as well as economic and other conditions which influence them.

We are not emitting, by and large, acids, though; we are emitting substances which are converted to acids, and there is a very complex chemistry involved here. We have considerable research going on in that and, in fact, Dr. Calvert will be discussing what his committee has done evaluating the status of our knowledge in that area.

But just let me say again that it is a complex set of chemistry. A lot remains unknown in the area, although again, we do know that these transformations take place and do lead to the acidic material being produced and deposited in a variety of forms.

In terms of the atmosphere, there is no question that the precursors to acid rain, the pollutants that give rise to it, are transported long distances. There is also no question that acid deposition in any given area is a combination of both the local sources and material transported long distance.

Where the uncertainty—I think the consensus of the scientific community is that we are not yet able to determine what amount of the acidity in a given area is from local versus long-distance sources, and maybe even more importantly, we are not able yet to have models with any certainty that can apportion how much a given source is contributing to a given sensitive region.

We are doing considerable work in this area. One of the things that I'd like to point out particularly is the so-called Captex experiment, the cross-Appalachian tracer experiment, which we are jointly doing with Canada and with involvement of the Electric Power Research Institute. This is an experiment where we are releasing this month and through the summer inert tracers in Ohio and up in Sudbury, Canada, and watching movement of this material in the atmosphere using aircraft and ground-monitoring stations to determine just where the pollutants eventually end up. It will certainly help us in verifying our models and understanding the movement of the pollutants, however it does not answer some of the critical chemical questions, and so further studies are going to be required to do that. But it is a good start and it is a major cooperative effort we are working with Canada on.

Acid rain, in and of itself, is a misnomer because actually we are talking about the deposition of acids in many forms. In fact, in many parts of the country, much more than half of the acids are arriving not in the wet form as snow or fog or rain, but in dry forms as gases and particles.

The problem we face here is that we can monitor the wet deposition quite well. We can determine the acidity of rain and so forth to quite some precision. We have no agreed-to method to measure these dry forms of deposition.

So in other words, a good portion of the problem of the acids reaching ecosystems and various sensitive resources is not even yet measurable in a way that we can agree to. We are putting extensive efforts into the development of technologies to do that; it will be some years, though, the scientists agree, before we have that capability.

In the area of wet deposition, we are implementing a major national trends network. We are working closer with the Canadians and being able, as of since about 1979, to produce maps such as this, which show the—this is again wet deposition—this is the acidity of rainfall, so there is nothing about the dry deposition, but looking at the distribution of the acid precipitation on a nationwide basis.

Again, as you can see from the map, the areas of highest concentrations of acids are in the northeast of Canada and the United States, and in fact, out in the West, some of the contours there probably should be dashed. There are very few sites and very little information out there, although we know in some local areas such as the LA basin and elsewhere, that very high acidic events occur. We are trying to improve. By next year, we will have in place—there are about 90 sites now, there will be about 150 sites in place. We will have a nationwide network quite adequate in the future to determine what trends occur and so forth.

Mr. OTTINGER. Mr. Chairman, could I ask a question at this point?

Mr. FUQUA. Sure.

Mr. OTTINGER. Where do these dry sources come from, the particles and the gas, and do you have any idea of the relative——

Dr. BERNABO. Yes, they come——

Mr. OTTINGER [continuing]. Contributions?

Dr. BERNABO [continuing]. From the same sources as the wet material. What happens is some of it—some of the dry material forms condensation, nuclei becomes entrapped within rainstorms and is brought to Earth in rain. Other cases, it just falls out dry like a dust in your house, if you will. So it is the same sources; it's not really the difference. It is whether it's brought to the ground by just settling out of the atmosphere in a dry form or whether it's involved in some precipitation event.

Mr. OTTINGER. Does it transport in the dry form the same way as it does in the wet form?

Dr. BERNABO. It does. The main difference is in the chemistry that occurs, whether it is occurring in a dry phase or wet phase, but we are not talking about different materials; we are just talking about different ways in which it is deposited.

Mr. OTTINGER. I see.

Thank you very much, Mr. Chairman.

Dr. BERNABO. In the case of the deposition monitoring, again, we are implementing a major network and one of the problems that I think you are all aware is the question arises, well, what has been the trends? Has acid rain been getting worse, has it been getting better, so forth?

The problem here is, and the consensus, I think, is pretty strong that we did not have in place before the late 1970s adequate monitoring data to really determine on a nationwide basis what trends were.

We hope very much, and we are putting in place the ability—that is not going to be the case in the future. As things change in the future, we will be able to determine that. It has been a source of much controversy because of inadequate data.

To give you an idea of why this is a problem, I think one of the things you have to understand about acid deposition is that it is a highly fluctuating variable. This is just some data showing changes in precipitation chemistry here in Washington, D.C. from 1976 until 1981. You can see it is averaged on seasons. You can see the great fluctuations by a factor of two or three between, let's say, the summer and fall—spring times—typically with the most acid precipitation occurring in this area in the summertime.

The reason I show this is to indicate two things: First thing is you don't just have a simple progression; you have a rather highly variable situation and in fact, this is one of the few sites where we have even this much data. There is only one place in the whole country where we have a record that is longer than a decade and that is in the Hubbard Brook experimental station up in New Hampshire, and that particular site where we do have good data back to the nineteen sixties, there is very little change in the acidity of rainfall over the last 20 years.

In some other sites here in Washington, one might look at that and say, well, it looks like things have become less acid; we really don't know why; I don't think it is a long enough record to firmly

state that, but again, the importance of understanding why you need a long history of monitoring is because this is a highly variable situation.

Also, even from the beginning to the end of a single rainstorm, you can have the variability of as much as 10- to 100-fold in the acidity of precipitation. It is a very strongly varying variable.

Now, I think in the area of effects is where some of the greatest misunderstanding or misinformation about acid rain exists. One must clearly distinguish, certainly for scientists, the difference between alleged or potential effects and effects which we actually know are occurring under ambient acid rain conditions such as the rainfall that we have today.

I am going to try to give you some indication of—it is certainly important for a policymaker to know both the potential threat, but it is also important to distinguish what the actual damages that we have documented are.

In this case, I think it's fair to say there is sort of a continuum between the areas where we are quite certain that acid rain is a major factor in influencing the various ecosystems to areas where we are much less certain.

In the case of aquatic systems, the strongest evidence exists and the consensus of the scientific community is specific sensitive aquatic resources have been impacted by acid deposition. In all the other areas, there is considerable controversy, and in fact, to my knowledge, there is no documented, out-in-the-field-under-ambient-conditions, effects widely accepted in any of the other areas, although there is very strong concern in each of them. That is to say, mechanisms have been identified, but the actual links have not yet been determined. I will discuss each of these a little more specifically.

In the case of aquatic resources, as I said, there's very strong agreement that this is an area where some impacts have taken place, but the question that remains is what is the scale of the resources at risk. What we tried to do for the policymakers is first determine what are the resources out there that are vulnerable; once you determine the ones that are at risk, then which of those have actually been impacted.

This map shows the areas of the country, based on actual surface water samples in streams and lakes throughout the country, the areas where the highest probability of these resources are being affected. In other words, where are the vulnerable resources?

Now, the areas of the—the very darkest areas are the areas of sensitivity. The lighter and yellow areas are quite the opposite; the areas that are not sensitive, they are insensitive to deposition of acids. You can see, again, things like the Adirondacks showing up in New York there, as one might expect. Many other regions where there are thin soils, high alpine areas such as in the Sierras and elsewhere, also showing up as potentially sensitive.

We are now going in and mapping these areas in much more detail to determine the finer structure and also look at—once you look at the population of what's sensitive, how many of those have actually been affected.

The most extensive work has been done in the Adirondacks and according to the State of New York about 4 percent of the surface

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area of the lakes—the so-called 200 lakes, if you want to look at it—the number of lakes that are about 2,500 lakes—are in critical condition and about 20 percent are vulnerable, which means roughly 75 percent are not sensitive under current situations.

So within these areas, again, is a great deal of variability in the sensitivity, but this does show the sensitive areas, so a significant portion, although not a large portion of the aquatic resource has been identified as being both sensitive and potentially impacted.

We are trying to extend again this now, so we are looking nationwide. We started up in the Northeast where the most concern is, but we are looking at all these areas and monitoring both not only the chemistry, but the biology to see what's happening in trends of the biologic populations.

One other area which I want to mention is we're also doing the Northeast. There is, of course, concern about drinking water supplies and we are in the process now of conducting specifically of drinking water sources—surface sources—an analysis of what the state of those resources are, to see how many of them and where the potentially affective ones are.

Now, beyond looking at what the scope of the resource at risk is, we, of course, have to understand the processes. One of the problems here is it is very complex; it is not just the matter of the rainfall coming into the lake. Most of the water which comes into many of these lakes first falls upon vegetation, trickles through the soil, and there can be as much as a 10- to a 100-fold difference in the acidity rainfall from the time it reaches the top of the forest to the time it actually trickles to the soil and enters the lake.

The role of the vegetation of the soil types is extremely important in determining the response of the lakes. In fact, in the ILWAS study sponsored by EPRI, it was found in the Adirondacks several lakes, only some miles apart, receiving the same acid rain responded entirely differently, some acidifying, some not, based on the difference in geology and so forth locally.

We are putting a lot of effort into developing models that will enable us to predict which of these lakes, in fact, will acidify and at what rates. But at this time, I think it is fair to say the consensus is we are not certain of what the rate at which the acidification is occurring.

We are also going back and looking at the sediments, material deposited as much as 100 or more years ago, to look at when these lakes became acidic and to try and determine from those sediments the rates at which this has occurred, because the existing data, in terms of surveys, is quite weak.

Moving on to the terrestrial area, one of the concerns, of course, is crops, and we have done a considerable amount of work in the laboratory and much of the work, in fact, is rather contradictory. Many of the studies show that some crops, in fact, do better with acid depositions, some do worse, depends on the condition. Sometimes even the same experiment on the same crop in two different universities have shown entirely different results.

Now, the reason for this, and the reason that some of the crops actually do better in acid deposition is quite simple. We are not dealing here with some toxic chemical like PCB or some—some-

thing which is foreign to ecosystems. We are dealing with sulfur and nitrogen, basic nutrients involved in fertilization of plants.

The rate at which these things are being deposited by acid deposition on croplands is considerably lower than the rate that farmers go out and intentionally apply through fertilizers. However, we are still concerned, and of course farmers manage the pH of their soils routinely through liming. But we are concerned about foliage and other effects, but we are particularly concerned about not so much what happens in the lab, but what happens in the real world where these plants are not only being acted upon by acid rain, but by ozone, by stresses of drought and other factors. Look at real-world situations.

So we are moving our emphasis more into the actual areas of production to assessing what the effects might be and looking primarily at the crops of major economic value, things like soybeans and corn, where if even a small reduction in yeild is, indeed, taking place, it would have major and important implications economically.

Beyond crops, though, I think there is a lot higher degree of concern for forests. Now, there are several reasons for this. First thing, forest lands are not as intensively managed, and secondly, unlike a crop which may only grow for a couple of months and be harvested, forest trees are exposed to acid deposition over periods of many decades and therefore can be more subject to cumulative and other effects.

At the current time, there is no question that forests are showing stresses—environmental stresses in a number of parts of the world. Germany and the forests—you are all familiar with the spruce dieback observations taking place in Northeast. No question that these changes are taking place.

What remains is the question to what extent acid rain, relative to ozone, relative to drought and a number of other factors, is causing these effects.

I recently attended a conference sponsored jointly by the U.S. Forest Service and Canadian Forest Service up in the city of Quebec where we had experts from around the world, including the Germans and the Swedes, as well as people from North America. It was a consensus of that scientific group that at the present time, it is not clear what role acid deposition is playing in affecting forests, however it was also the consensus that it is a primary suspect in a number of the changes taking place. There is very hot scientific debate right now in Germany as to what extent acid deposition versus the direct fumigation by ozone and other pollutants are responsible for the degredation of those forests which can be observed.

Now, I might point out that in looking at the effects of acid deposition on forests, this is a very new area of investigation. However, we have been studying the effects of other air pollutants, notably ozone and oxidants, on forests for decades, and there is no question that those air pollutants, particularly the oxidants, have major impacts on both crops and forests, so we clearly have some other pollutants where we have a much better indication.

For instance, in California, a number of the agricultural and other crops no longer can grow downwind of Los Angeles, specifi-

cally due to the high levels of these oxidants. So we are dealing with the situation where for some air pollutants, we already know that they are major factors; the question for us is what—what extent is acid deposition contributing to those observed changes?

Some of the researchers believe in 2 to 3 years, we will have a much better idea of that; others are less optimistic and feel because of the complexities of the interaction of these various pollutants, things such as drought and pests, it may be harder to sort it out, but we are putting major emphasis because we feel that the potential here is quite great in terms of the role of acid deposition.

And I might say, it is not only through the plants themselves and direct effect on the needles and other portions, but through the soil. The soil mechanism here could be very important in determining the response of these organisms to acid deposition.

Now, one other thing, just to point out, is in Sweden, where much of the forests are very limited in nitrogen, the forests have shown in recent years more rapid growth, and it is widely believed now that that, in fact, is a positive effect, if you will, of acid deposition. That's not to say acid deposition is good, but again, it points out that we are dealing here with basic nutrients; it's not a simple matter of a foreign substance injuring the things. You have to balance both the positive and negative responses of the plants.

Another concern, of course, is materials and there are major concentrations of materials, particularly in urban areas, but elsewhere. Sensitive things like limestone and concrete and so forth. We are conducting a variety of exposure studies, exposing materials in different areas receiving different levels of acid deposition, both wet and dry, and we are conducting in Northeastern United States a major inventory examining what is the size of the material resource in Northeast at risk. How many of the materials are at risk?

There is no question that higher acidity leads to a greater rate of corrosion and erosion of these materials; the question is, What role does acid deposition play relative to other pollutants, particularly in the urban environment and normal weathering processes? So we are inventorying again the size of the resource at risk and we are not only concerned here with the common materials, such as used in modern buildings, but also a variety of cultural resources—in this case the Brooklyn Bridge or things like the Statue of Liberty and so forth, major things which their value cannot just be measured in economic terms, which you may want to obviously protect and preserve and have some sense of whether they, indeed, are threatened and to what degree by the deposition.

I've got a bulky projector.

In the area of control technologies, our program is not developing control technology hardware. That is done under a number of other programs. The budget the Federal Government, EPA, and Department of Energy spend on that far exceeds the budget that we spend in acid rain research. What we are doing is examining the control technologies that are out there that are relevant to acid rain.

Now, the reason for this is that there is no control technology for acid rain. There are control technologies for the precursors to acid rain, but there are a number of other reasons beside acid deposi-

tion. For instance, one might be concerned about controlling sulfur dioxide emissions, things like visibility, possible health effects.

So what we are doing—this is an analysis. We are particularly concerned here; we are looking mostly at sulfur dioxide, although I caution you to understand that there is no question at all that nitrous oxides are a contributor, but we are focusing mainly on the sulfur dioxides since those are the primary sources in the Northeast, and we are looking at the retrofittable technologies because the new plants that are going to come on-line and the utilities and elsewhere, meeting the new source performance standards, will, in fact, be very clean. It is the older plants which we're worried about.

So we are looking at technologies that are retrofittable. We're examining the ability of them to reduce the emissions and their costs and so forth, their applicability, the number of work going on in here.

Probably the most interesting one is the so-called LIMB, the limestone multistage burner, where you can reduce both sulfur and nitrogen emissions by about 50 percent. That is in the prototype developmental stage and we look forward to that as a—it's a—I think it's about 50 percent to 75 percent cheaper than scrubbers and is retrofittable into older plants. So we see that as a promising potential technology.

Finally, when you look at all these various components, you have to pull them together, certainly for people like the Congress and the regulatory officials in the administration elsewhere, into what does it all mean? What are the risks involved? What are the damages? How do you trade that off against the possible costs and different mitigation strategies?

And so we have a group looking to try and assess, to pull together these various pieces of information, put them in a form that can be used for assessing the state of the situation.

We have a number, then, of special assessments coming out. I'll just—there is a long list of these—I'll just mention some. One of the, I think, least—one that's gotten some of the least publicity is one of the most important—is the so-called critical assessment document. Now, this is a document which is produced by 54 scientists from around the world, each experts in their areas—it has been widely reviewed by the public and other scientists—where they are each asked to examine in their field of expertise specifically what is the state of the science. This is undergoing final review now and will be published fairly soon. So it is sort of a benchmark by a wide range of scientists outside of the policy realm of where we stand in the science.

We have also developed, as I say, a monitoring strategy. The assessment of the sensitive lakes and streams will be completed this year so we will know what the extent of the resource is out there. We are looking at trends, a number of other factors.

Besides these specific assessments, we at some points are going to do what we call integrated assessments, that is, pull together, when a set of a critical mass information exists, and make statements larger than just on different parts of the problem.

The first of these is now underway. We're calling it our 1985 assessment and it's basically a damage assessment. We believe the

state of the science is such that in 1985, we will be able to state, with some limits of confidence, but we will be able to make the first relatively definitive statement of what the scale of damages to aquatic, terrestial, and other systems are in the country.

We will also, along with that, quantify the level of uncertainties that exist in the other areas. It, however, will be some time beyond that, just in terms of the science and the rate at which the science is progressing, before we will be able to really provide the kind of source receptor relationship information that would be required to do scientifically based optimizations of control strategies.

But nonetheless, we think it is very important, regardless of what policy's involved, to have some sense of what is the size of the resource at risk, how much damage has occurred and what are we really talking about in terms of what we're trying to protect.

[See slide 6, p. 25.]

Let me say a little bit about funding of the program. In 1981, prior to the initiation of this congressionally mandated program, there were piecemeal efforts throughout the Federal Government——

Mr. FUQUA. Dr. Bernabo——

Dr. BERNABO. Yes.

Mr. FUQUA. We have a vote and I think it might be appropriate right at this point to take a break so we can run out to vote and we will come back and finish your statement.

Dr. BERNABO. Certainly.

Mr. FUQUA. The committee will stand in recess.

[Recess.]

Mr. FUQUA. The committee will resume. It looks like we've lost our witness. [Laughter.]

Mr. FUQUA. Oh, there he is. Please resume.

Dr. BERNABO. Yes, OK.

[See slide 7, p. 26.]

I've been discussing the budget, the funding which has gone into the federally sponsored research which is, again, done in a wide variety of areas. Prior to the initiation of this national program, there was roughly $11 million in piecemeal efforts done by the Federal agencies.

[See slide 8, p. 27.]

In 1983, the program had doubled and begun an integrated program at about $22 million. The request before Congress now is for a further 24-percent increase to about $27 million.

[See slide 9, p. 28.]

So there has been a substantial increase in this funding, enabling us to begin filling some of the gaps and trying to accelerate the effort.

[See slide 10, p. 29.]

Now, this particular diagram shows for the current fiscal year just ending this month the distribution of those resources. In other words, where are the resources targeted in terms of the research? As you can see, the large chunks are in the areas that we have major concerns, like terrestial impacts and aquatic impacts. Much of the aquatic work is, at this point in the program, surveying. In the future years, we plan to shift the efforts, once we have the surveying work done, much more into projections and modeling work.

In the area of deposition monitoring, that, of course, is, as I said, developing the technologies to monitor dry depositions so that in the future we could be—get a handle on that whole aspect of the problem and implementing and maintaining a long-term national network.

We also maintain, I might say, a global network. We have sites in different parts of the world looking again at what the background globally is, places like Hawaii, Amsterdam Island in the southern Indian Ocean and Australia.

In the area of atmospheric processes, again, considerable controversy and need for study in this area, these things like the CAPTEX experiment and the chemistry work, all included in that, and then smaller amounts of money going into the areas like man-made natural sources, we're primarily doing inventorying and modeling work and some effort in the assessment area.

We expect the assessment portion of the budget to grow in future years as the database improves and allows us to make more cogent assessments and then some work on the effects of materials. Now, the materials there actually is doubled in the last few years, although it is still a small amount of money, very little research had been done in that area.

[See slide 11, p. 30.]

So that's how the dollars are distributed at the current time, some sense of who provides those dollars, while there are 12 agencies in the program, 5 of them make up the lion's share of the work, the Environmental Protection Agency currently funding about half of the program and then the roughly equal shares between the other major players which are Agriculture, NOAA, Interior, and Energy.

Now, there is a substantial pot of money, if you will, out there also at the National Science Foundation. Roughly $1.5 million a year are going into acid rain research. We do not count that in our budget. The reason is we feel that that sort of a reservoir of money out there that doesn't have to pass the relevancy test. All this money—all these projects have to be directly relevant to answering the questions we feel are important about acid rain.

Of course, as you know, there may be a number of very good ideas that we don't see the relevance of right now, but 5 years from now, may turn out to be actually have been the best questions, so we have a reservoir of money out there that the National Science Foundation is a direct part of this program. We do not include them in the rigorous budget planning; we leave them out there just to fund the best science based on merit, and not any other criteria.

So this is a—if you will, it's a conservative estimate of the money we're spending on acid rain. Also, there is considerable spending on a variety of other things that we're not counting in here; that is, basic ecology research, which—and atmospheric process research, which lays the groundwork for our work.

[See slide 12, p. 31.]

In summary, then, let me say something about the strategy of the program and then conclude. The first thing, as I said, there was existing efforts, not only here, but they have been studying this issue for decades in Scandinavia. The Canadians have a major

research program as large as our own, and we are building upon those previous efforts, not reinventing the wheel.

We are using the existing data and knowledge, the overview I gave you of the science isn't just work that we sponsored; it's work that private-sector groups and environmental groups of other nations have sponsored. We're trying to work with the consensus, objectively look at what's available throughout the scientific community.

We feel that this establishment of a long-term monitoring network is crucial, so that as we go forward in the future and economic conditions change, emissions change due to control strategies, that we don't have the problems we've had in the past of determining what did we get for those changes; what has happened. So this is a, we think, a fundamental thing. If we had this kind of long-term monitoring before this issue became so charged, we would, in fact, be in much better shape today to assess where we stand.

And finally—probably most importantly, we see the purpose of the research we're conducting is not just to do a lot of nice research and keep scientists employed, but in fact, to do the research most relevant to the kinds of questions policymakers are asking, and we try and provide a very rigorous test of that by working with specific objectives and making each of the programs—each of the projects—responsive to objectives.

One of the ways we keep this on track is we have sponsored a number of reviews. Outside experts—we just had one up in Boston recently—where all of the atmospheric research in deposition monitoring and sources research—each and every principal investigator was reviewed, not only for the scientific quality of all those projects, but also for their relevance to our goals and the University Corporation for Atmospheric Research sponsored that research for us.

Back in the fall, we did a similar review with outside experts from around the world examining our effects research and that was conducted for us by North Carolina State. So we have had a number of outside groups helping us examine the quality of the science and its relevance to the key questions.

And finally, we are periodically assessing the information. We're not going to wait some mythical time in the future when all the answers are available; clearly there'll always be uncertainties. We see ourselves as progressively reducing those uncertainties and providing the information in the most timely manner to the policymakers, to the public, and other interested parties.

So if I might conclude then, I think that there has been a very broad progress made during the first year of the national program. We have laid the framework for future years for attaining much of the information that will be useful. I think it—we are trying to focus more and more the research on the key issues that are going to be of most importance to the policymakers and that as we do our current assessments, we will be able to not only tune the research relevant to what the—what we're learning, but also tune the research relevant to what actions the Congress, the administration, so forth, may take.

It is clear at this point that the science can provide some guidance, but we're not in a position to make a straight-out, based-on-

science recommendation of here's the prescription and we know it will work for what to do.

What we see ourselves as doing, then, is providing an increasingly better information base. The difficult decisions, really, are ones as much in the realm of politics as in science.

Thank you very much.

Slide 1

ACID PRECIPITATION ACT OF 1980

PROVISIONS

1. ESTABLISHED INTERAGENCY TASK FORCE ON ACID PRECIPITATION

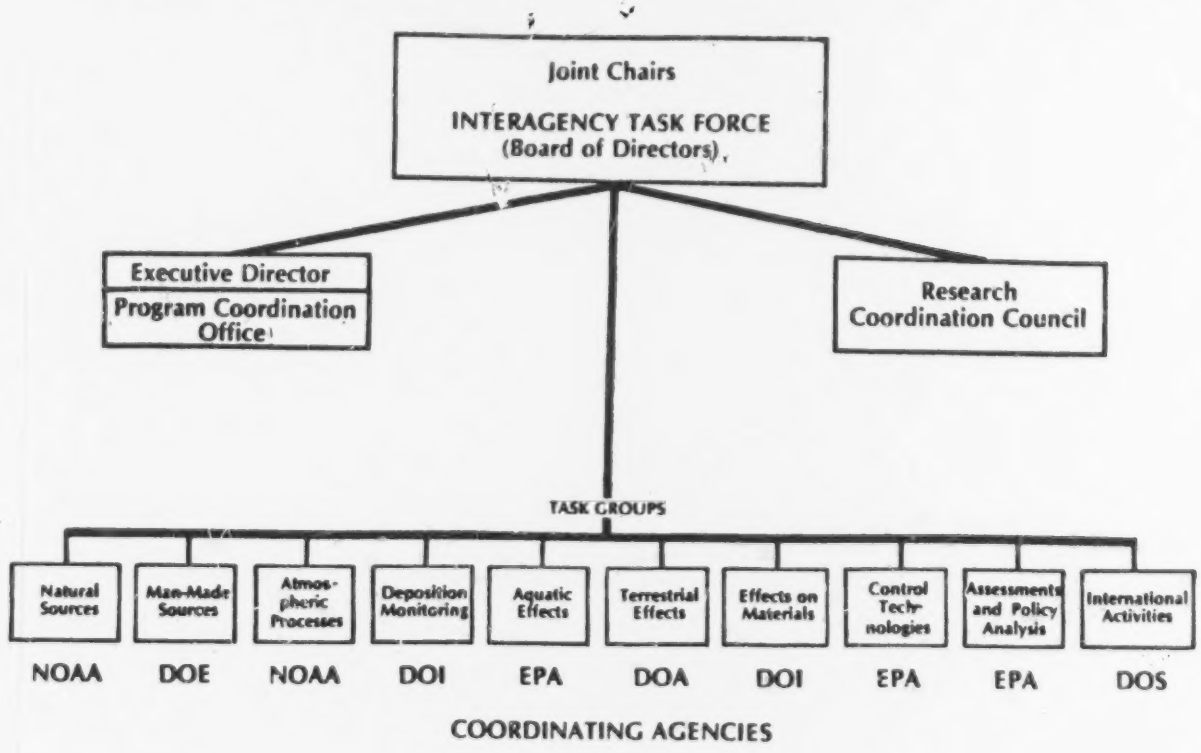
   - JOINTLY CHAIRED BY EPA, DOA, NOAA
   - INCLUDES: CEQ, DOE, DOI, DHHS, NASA, NSF, TVA, DOS
     ALSO: ARGONNE, BROOKHAVEN, OAK RIDGE, PACIFIC NORTHWEST NATIONAL LABS.

2. AUTHORIZES $5M/YEAR FOR 10 YEARS TO NOAA

   - NOAA - DIRECTOR OF RESEARCH
   - NATIONAL LABS TO FORM "RESEARCH MANAGEMENT CONSORTIUM"

Slide 2

National Acid Precipitation Assessment Program



Slide 3

RESEARCH CATEGORY	COORDINATING AGENCY
A. Natural Sources	NOAA
B. Man-made Sources	DOE
C. Atmospheric Processes	NOAA
D. Deposition Monitoring	DOI
E. Aquatic Effects	EPA
F. Terrestrial Effects	DOA
G. Effects on Materials	DOI
H. Control Technologies	EPA
I. Assessments and Policy Analysis	EPA
J. International Activities	DOS

Slide 4

TASK FORCE RESPONSIBILITIES

1. DEVELOP AND UPDATE TEN-YEAR NATIONAL ACID PRECIPITATION ASSESSMENT PLAN

2. IMPLEMENT AND OVERSEE A COMPREHENSIVE NATIONAL PROGRAM

   - COORDINATING FEDERAL AGENCIES ACTIVITIES
   - DEVELOPING ANNUAL INTERAGENCY BUDGET
   - MAINTAINING INVENTORY OF FEDERALLY-SPONSORED RESEARCH
   - ENCOURAGING PRODUCTIVE INTERACTION WITH NON-FEDERAL ACTIVITIES

Slide 5

TASK FORCE RESPONSIBILITIES

3. PRODUCE ANNUAL REPORTS ON PROGRAM'S PROGRESS AND POLICY IMPLICATIONS

   - SPONSOR ANNUAL WORKSHOP AND PERIODIC ASSESSMENTS

4. ENSURE U.S. NATIONAL PROGRAM IS SUPPORTIVE OF, AND COORDINATED WITH, RELEVANT INTERNATIONAL ACTIVITIES

Slide 6

BUDGET PROCESS

- TASK FORCE PROVIDES GUIDANCE TO AGENCIES ON THE IMPLICATIONS OF THE CURRENT PLAN FOR THEIR PROGRAMS
- OMB ADVISES TASK FORCE OF TARGET LEVELS FOR TOTAL PROGRAM
- TASK FORCE DEVELOPS INTERAGENCY BUDGET RECOMMENDATIONS
- AGENCY BUDGET REQUESTS ARE REVIEWED BY OMB AS PART OF AN INTEGRATED MULTIAGENCY PROGRAM

Slide 7

NATIONAL ACID PRECIPITATION ASSESSMENT PROGRAM

ACTUAL EXPENDITURES
FY 1982
($ in thousands)

	EPA	DOA SE	DOA FS	NOAA	DOE	DOI PS	DOI GS	DOI FW	TVA	TOTAL
A. Natural Sources				600						$ 600
B. Man-made Sources	870				300					1170
C. Atmospheric Processes	3273			600	805	50			126	4854
D. Deposition Monitoring	797	253	127	700	497	75	585			3034
E. Aquatic Effects	1475		135		560	94	580	75	133	3052
F. Terrestrial Effects	1355	264	570		122	178	295		66	2850
G. Effects on Materials	250					178				428
H. Control Technologies	(10,400)[a]				(6200)[a]					(16,600)[a]
I. Assessments and Policy Analysis	1105				260					1365
Subtotals:		517	832			575	1460	75		
TOTALS:	$ 9125	1349		1900	2544	2110			325	$ 17,353

[a] Control Technologies figures not included in total. These funds for general development of SO_2 and NO_x control hardware are appropriated under other preexisting programs. The Control Technologies Task Group coordinates these efforts with the assessment and research activities of the National Program.

Slide 8

NATIONAL ACID PRECIPITATION ASSESSMENT PROGRAM

FY 1983
($ in thousands)

	EPA	DOA SE	DOA FS	NOAA	DOE	DOI PS	DOI GS	DOI FW	TOTAL
A. Natural Sources				700					$ 700
B. Man-made Sources	1050				300				1350
C. Atmospheric Processes	3858			1050	274	50			5232
D. Deposition Monitoring	1145	404	145	500	666	100	1449		4409
E. Aquatic Effects	1935		210		550	148	445	75	3363
F. Terrestrial Effects	1468	946	1258		50	229	486		4437
G. Effects on Materials	410					535	50		995
H. Control Technologies	(7600)*				(6500)*				(14,100)*
I. Assessments and Policy Analysis	1570				220				1790
Subtotals:		1350	1613			1062	2430	75	
TOTALS:	$ 11,436	2963		2250	2060		3567		$ 22,276

* Control Technologies figures not included in total. These funds for general development of SO_2 and NO_x control hardware are appropriated under other preexisting programs. The Control Technologies Task Group coordinates these efforts with the assessment and research activities of the National Program.

Slide 9

NATIONAL ACID PRECIPITATION ASSESSMENT PROGRAM

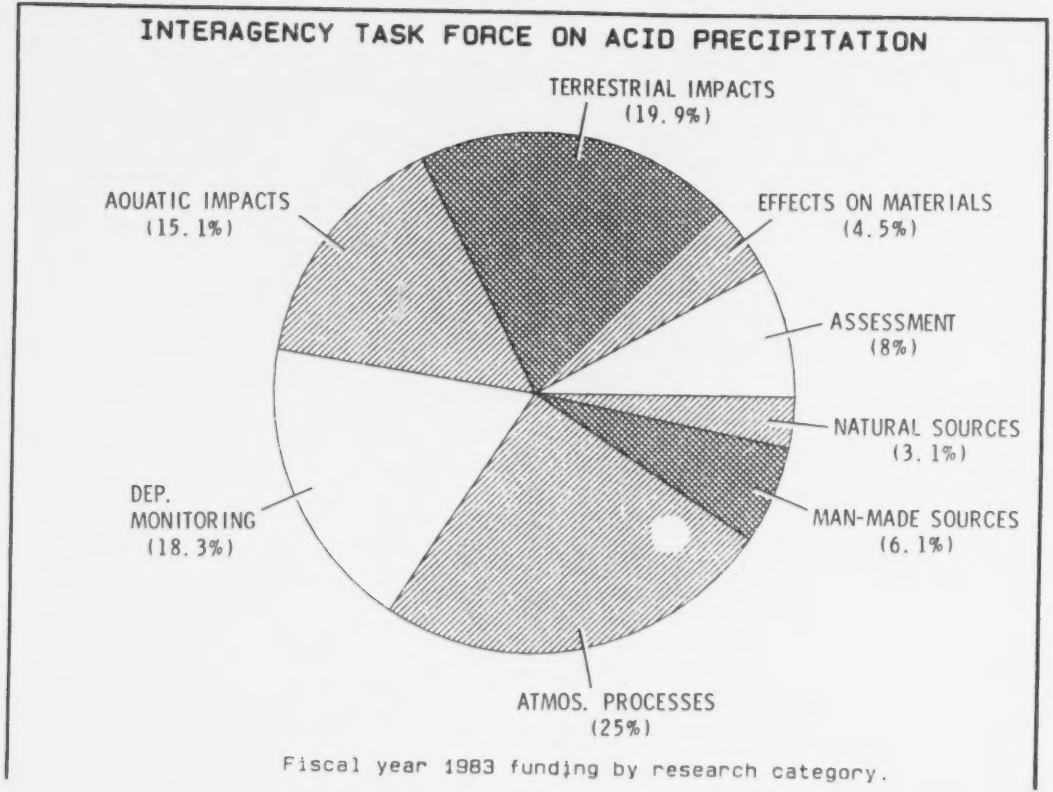
Proposed revisions to the President's Budget Request (May 1983)

FY 1984
($ in thousands)

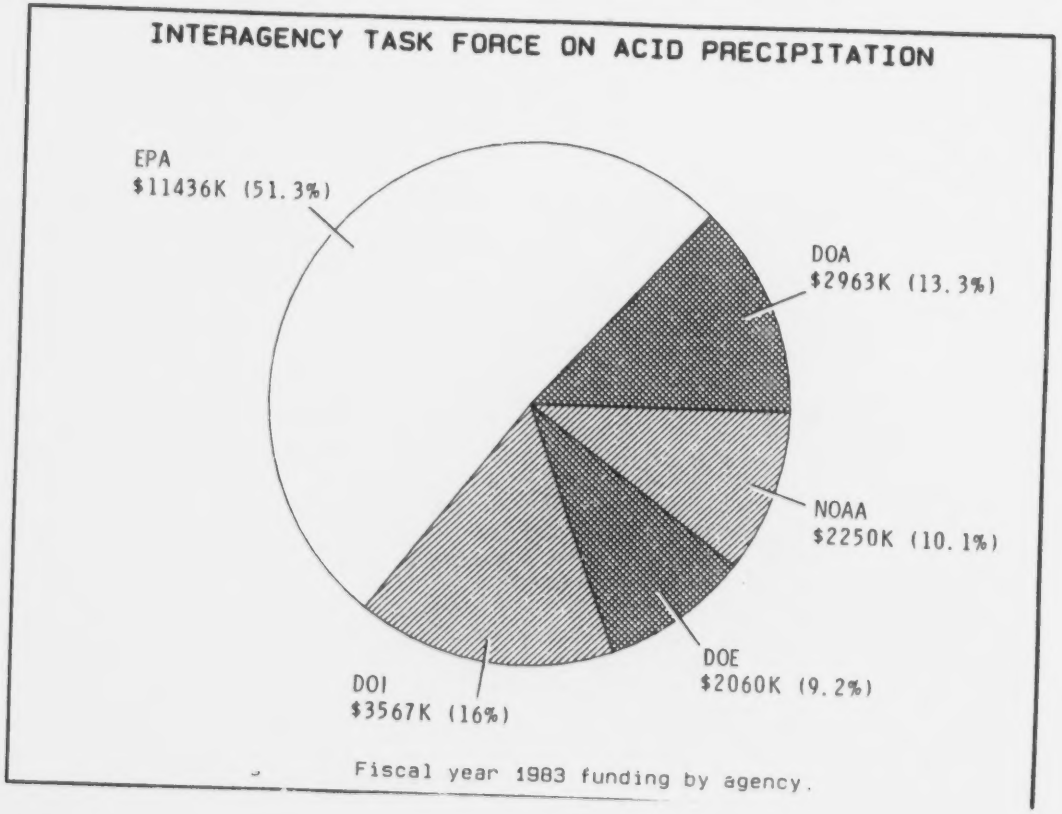
	EPA	DOA SE	DOA FS	NOAA	DOE	DOI PS	DOI GS	DOI FW	DOI BLM	TOTAL
A. Natural Sources				955						$ 955
B. Man-made Sources	1090				260					1350
C. Atmospheric Processes	5250			1020	270					6540
D. Deposition Monitoring	1789	274	146	1130	942	122	1870		80	6353
E. Aquatic Effects	2285		210		550	148	445	125		3763
F. Terrestrial Effects	1468	946	1258		50	229	486			4437
G. Effects on Materials	605					740	150			1495
H. Control Technologies	(4100) *				(7000) *					(11,100) *
I. Assessments and Policy Analysis	1500				1175					2675
Subtotals:		1220	1614			1239	2951	125	80	
TOTALS:	$ 13,987	2834		3105	3247		4395			$ 27,568

* Control Technolgies figures are not included in totals. These funds for general development of SO_2 and NO_x control hardware are appropriated under other pre-existing programs. The Control Technologies Task Group coordinates these efforts with the research and assessment activities of the National Program.

Slide 10


INTERAGENCY TASK FORCE ON ACID PRECIPITATION
TERRESTRIAL IMPACTS
(19.9%)
EFFECTS ON MATERIALS
(4.5%)
ASSESSMENT
(8%)
NATURAL SOURCES
(3.1%)
MAN-MADE SOURCES
(6.1%)
ATMOS. PROCESSES
(25%)
DEP.
MONITORING
(18.3%)
AQUATIC IMPACTS
(15.1%)
Fiscal year 1983 funding by research category.

Slide 11


INTERAGENCY TASK FORCE ON ACID PRECIPITATION
EPA
$11436K (51.3%)
DOA
$2963K (13.3%)
NOAA
$2250K (10.1%)
DOE
$2060K (9.2%)
DOI
$3567K (16%)
Fiscal year 1983 funding by agency.

Slide 12

PROGRAM STRATEGY

- BUILD UPON PREVIOUS EFFORTS
- USE EXISTING DATA AND KNOWLEDGE
- ESTABLISH A LONG-TERM MONITORING NETWORK
- CONDUCT NEW RESEARCH TO DEVELOP KNOWLEDGE NECESSARY FOR POLICY DECISIONS
- PERIODICALLY EVALUATE INFORMATION AND ASSESS ITS IMPLICATIONS

[The prepared statement of Dr. Bernabo follows:]

STATEMENT OF

DR. J. CHRISTOPHER BERNABO
EXECUTIVE DIRECTOR
NATIONAL ACID PRECIPITATION ASSESSMENT PROGRAM

BEFORE THE
SUBCOMMITTEE ON ENERGY DEVELOPMENT AND APPLICATIONS
AND THE
SUBCOMMITTEE ON NATURAL RESOURCES, AGRICULTURE RESEARCH AND ENVIRONMENT
OF THE
COMMITTEE ON SCIENCE AND TECHNOLOGY
UNITED STATES HOUSE OF REPRESENTATIVES

13 September 1983

Mr. Chairman, other Members and guests:

It is a pleasure to be here to describe the statutory National Acid Precipitation Assessment Program and our recent annual report to the President and Congress. I serve as the Executive Director of the National Program while on assignment from the National Oceanic and Atmospheric Administration (NOAA) Administrator's Office of Policy and Planning.

The purpose of the National Acid Precipitation Assessment Program is to successively improve our understanding of the causes, effects and possible solutions to the acid rain problem. The Program includes research, monitoring and assessment activities that emphasize the timely development of a progressively firmer scientific basis for decision making by the Congress, regulatory agencies, private sector, environmental groups, and the public. The National Program involves over 200 projects and hundreds of scientists in government, academia, and the private sector.

My testimony today will include descriptions of the authorizing legislation, the organization and implementation of the Federal acid rain research effort, and a synopsis of the major research findings and accomplishments of the National Program's first year of research.

LEGISLATION AND ORGANIZATION

The Interagency Task Force on Acid Precipitation was established by the Acid Precipitation Act of 1980 (Title VII of the Energy Security Act - P.L. 96-294) to oversee the planning and implementation of the National Acid Precipitation Assessment Program. The Task Force is jointly chaired by the Environmental Protection Agency (EPA), the Department of Agriculture (USDA), and the National Oceanic and Atmospheric Administration (NOAA). The other members include one high-level representative from each of the twelve Federal entities in the Program: the Departments of Energy (DOE), Interior (DOI), Commerce (DOC), Health and Human Services (HHS), and State (DOS); the Council on Environmental Quality (CEQ); the National Aeronautics and Space Administration (NASA); the National Science Foundation (NSF); and the Tennessee Valley Authority (TVA). The Task Force also includes the directors of Argonne, Brookhaven, Oak Ridge and Pacific Northwest National Laboratories, as well as four Presidential appointees.

The primary responsibilities to the Task Force are:

- o Plan and manage the National Acid Precipitation Assessment Program.
- o Provide annual reports on the National Program's progress.
- o Produce an annual interagency budget for the National Program.
- o Coordinate the National Program with the research and monitoring activities of the private sector groups, environmental organizations, states, and other nations.
- o Maintain an inventory of Federally-funded acid deposition research projects.
- o Disseminate research results and assessments of their implications.

The role of the Task Force in planning the interagency budget for the National Program is a highly effective and unique aspect of the Federal effort. By working together through the Task Force, the agencies have established a research program that addresses national needs while building on the research expertise of the individual agencies. This strong interagency planning process avoids unnecessary duplication and eliminates crucial omissions in the National Program.

The Task Force itself functions as the "board of directors" for the National Program by setting the research goals, identifying the projects needed to meet these goals, and deciding which agencies are best suited to conduct the necessary work. This interagency planning process ensures an integrated and comprehensive program, with each agency contributing to specific aspects of the overall national effort.

The Task Force has 10 working-level Task Groups, one for each of the National Program's nine research categories and one for international activities. These technical groups include program managers and experts from all the participating Federal agencies and National Laboratories. They are responsible for the detailed planning and research in their assigned areas. The Task Groups with their Coordinating Agencies are:

Task Group	Coordinating Agency
A. Natural Sources	NOAA
B. Man-made Sources	DOE
C. Atmospheric Processes	NOAA
D. Deposition Monitoring	DOI
E. Aquatic Effects	EPA
F. Terrestrial Effects	USDA
G. Effects on Materials and Cultural Resources	DOI
H. Control Technologies	EPA
I. Assessments and Policy Analysis	EPA
J. International Activities	DOS

The Research Coordination Council (RCC) oversees and integrates the efforts of the various Task Groups and develops draft reports, program plans, budgets and other recommendations for consideration by the full Task Force. The joint chairs designated the Task Force's executive director to chair it. The RCC includes leaders of all the Task Groups, the chairperson of the National Laboratory Consortium, and other appropriate agency representatives.

The Task Force is vigorously implementing the comprehensive national research program. Federally-funded acid deposition research has doubled from about $11 million in FY 1980 to $22.3 million for FY 1983, and the President's budget for FY 1984 requests a further 24% increase in funding. The National Program is producing results at an increasing rate, and disseminating them as soon as they become available.

FY 1982 ANNUAL REPORT

In June of this year the National Acid Precipitation Assessment Program issued the Annual Report to the President and Congress on its first full year of research. The report summarizes the status of scientific knowledge, the major research findings and accomplishments of the National Program's first year and the outlook for continued progress. As the National Program progresses, future annual reports will provide updates and increasingly better information to assist decisionmakers and the public in continuing to resolve the acid rain issue.

STATE OF THE SCIENCE

This overview on the current state of the science is based on information from all available sources, including research supported by Federal agencies, States, environmental organizations, private sector groups, and other nations.

Causes

Man-made atmospheric pollutants are the major contributors to acid deposition in northeastern North America. The approximate coincidence of the region of highest precipitation acidity with the areas of greatest sulfur dioxide and nitrogen oxides emissions provides circumstantial evidence to support this assertion. This conclusion is also supported by preliminary meteorological studies tracking the physical movement of one pollutant (sulfur dioxide) from sources of emission to suspected areas of deposition. The National Program is also studying another contributory pollutant (oxides of nitrogen) and other chemical species suspected of being factors in controlling acid deposition. Current data and available methods, however, are not sufficient to establish the quantitative relationship between pollutant emissions and acid deposition on a regional scale, or under varying climatic conditions. It is not yet possible to identify the specific change in the acid deposition patterns that would result from a given change in presursor emissions.

In addition to man-made sources, natural sources of acid precursors are known to exist. On a global scale, it is roughly estimated that about half the sulfur in the atmosphere is from natural emissions. Regionally, preliminary findings suggest that natural sources might contribute significantly to the acidity of precipitation in some areas, such as the southeastern United States. In the northeastern United States natural sources are believed to be minor contributors to acid deposition. The National Program includes research to increase our meager knowledge of the strengths, character and distribution of natural sources of sulfur and nitrogen species that can contribute to acidity as well as alkaline dusts that neutralize acids in precipitation. This information will be used to assess the potential significance of these sources in contributing to acid deposition in sensitive areas.

Source/Receptor Relationships

To formulate effective control strategies, the relationship between emission of precursor pollutants and the deposition of acidic material at sensitive receptors should be reliably established.

The ability of current generation models to predict source/receptor relationships with adequate resolution and accuracy is an unresolved issue. A study on the ability of eight models to predict the deposition of sulfur emissions in eastern North America found only a few suitable monitoring data points. While a method is now available to properly evaluate model performance, more data (quality assured and available as a time series) is needed to quantitatively determine the uncertainties in the model predictions. In addition, there are widely differing views of the relative importance of "local sources" (acid deposition from nearby sources of pollution) and "long-range transport" (acid deposition from far-removed sources of pollution) on the receptors in the local region.

Because of the urgent need to improve our predictive ability to the point where we can reasonably estimate the changes in amounts of acid deposition expected from decreases in pollution levels, the National Program includes several simultaneous approaches: (1) for near-term assessments, modifying current-generation models to refine their accommodation of chemical and physical factors over shorter periods of time; (2) constructing a new generation of models to more effectively treat the physical movement of pollutants in the atmosphere, as well as the mechanisms for their conversion to acids and deposition; (3) designing a series of field experiments to provide empirical, quantitative evidence of the pollution source/acid deposition relationship; and (4) upgrading and evaluating a suitable local/mesoscale model to quantify the relative contribution of local versus long-range transport on receptors within the mesoscale region from a source or source area (up to 250 km) and to determine the importance of emissions of primary sulfates on acid deposition in this same region. Intensive efforts underway in the National Program and elsewhere should yield a more adequate ability to project the consequences of changes in emission patterns.

Impacts

Among the possible effects of acid deposition, aquatic effects are currently of greatest concern because of the sensitivity of certain surface waters to acidification and the known potential for damage to fish and other organisms. Initial surveys indicate that a small fraction of the lakes and streams concentrated in the sensitive regions of northeastern North America have been acidified, and their biota have changed. In the Adirondacks, one of the most sensitive regions in North America, the State of New York has classified 3.6 percent of lake acreage as in "critical" condition and 14 percent endangered, with 84 percent currently surveyed. Acid deposition is probably the major contributor to this acidification, but the rates of change and the factors controlling it are still poorly understood. Many of the reported cases that suggest acid deposition is the cause of some observed change in an aquatic ecosystem are based on circumstantial evidence and lack documentation of the mechanisms linking cause and effect. At present, the extent of actual aquatic damages is not well established. The question of long-term trends in freshwater acidification also remains uncertain.

The National Program produced maps in FY 1982 that depict the regions in which surface waters could be most susceptible to acidification. Studies are underway to better define the factors that control acidification, and improved regional monitoring and testing programs will produce more detailed and reliable inventories of the extent of damages and of the location and value of the resources at risk.

Beyond the alteration of the chemistry and biology of certain sensitive surface waters, the extent of other effects of acid deposition in North America are undetermined. There is concern about the possibility of effects on crops, forests, wetlands, soils, building materials, and indirectly to human health (through drinking water or food). Although mechanisms for such effects are known, it has not yet been demonstrated that they are widely occurring under ambient acid deposition. For instance, although experimental studies with simulated acid rain have shown both positive and negative effects on crop species, effects on crops growing under actual field conditions have not been established.

Similarly, although long-term declines in high-elevation spruce are observed in New England, the relative importance of acid deposition versus other factors such as drought, disease and other pollutants is not yet documented. In contrast to acid deposition, the effects of ozone and other air pollutants on crops and forests are well documented. The National Program is speeding up investigations and analyses in order to determine the actual effects of acid deposition.

NATIONAL PROGRAM FY 1982 ACCOMPLISHMENTS

Some selected highlights of progress in FY 1982 for the National Program's research areas are as follows:

A. Natural Sources:

o Developed and tested instruments for reliable field measurement of sulfur and nitrogen emissions from natural sources.

o Obtained preliminary measurements of natural sulfur compounds in the ocean that suggest that emissions from marine sources may contribute significantly to atmospheric acidity in some areas.

B. Man-made Sources:

o Completed preliminary study on emissions sources which suggested that nearby petroleum combustion, as well as local and distant coal combustion, could significantly affect local acid deposition.

o Continued development of models to predict the cost and emissions changes from utilities and industries based on various possible control strategies.

o Developed specifications for emissions inventories with improved spatial, temporal, and source-type resolution, and initiated studies to better characterize the man-made sources of acid precursor pollutants.

C. Atmospheric Processes:

o Obtained initial observations that confirm the importance of clouds in transforming sulfur dioxide to active sulfates.

o Completed planning and initial testing for a major field experiment to be performed in FY 1983 that will track the movement of tracer gases released in the midwest over hundreds of miles.

o Improved atmospheric models and applied them to gain a better understanding of the movement and conversion of pollutants and the deposition of acidic material.

o Initiated field studies that suggest the importance of man-made aerosol in cloud formation.

D. Deposition Monitoring:

o Produced the first comprehensive maps describing the distribution of major chemical species over North America.

o Completed design for the National Trends Network, which will be established in FY 1983 building on existing sites that meet specific criteria.

o Established four research sites to test improved methods of monitoring wet and dry deposition.

o Compared field measurements of dry deposition obtained from different prototype methods and continued effort to develop reliable techniques for routine measurement of dry deposition.

E. Aquatic Effects:

o Produced a nationwide map indicating regions whose surface waters are likely to be most sensitive to acidification.

o Began long-term monitoring of aquatic resources (chemistry and biota) in key sensitive regions.

o Completed preliminary survey of drinking water in the Northeast that indicates some reservoirs and their watersheds have limited ability to buffer acid deposition.

o Developed improved understanding of factors controlling the susceptibility of surface to acidification and evaluated models for predicting acidification.

o Evaluated existing information on mitigation strategies (liming), and initiated field and laboratory studies to examine their effectiveness and limitations.

o Began studies of acid deposition effects in the southeastern United States.

F. Terrestrial Effects:

o Initiated extensive long-term studies to determine whether acid rain has caused changes in rate of growth and species composition in forests.

o Completed a study evaluating whether acid rain predisposes pine seedlings to stress factors. It did not show an increase in susceptibility to insect or disease attack; some seedlings showed increased resistance to disease.

o Conducted agricultural experiments using simulated acid rain that did not indicate injury to potato or two forage plants, but decreased the yield in soybeans and corn under some conditions and had no effect under others.

o Performed soil studies in the Southeast, mid-Atlantic coast, and Northeast that showed variable response to acid rain relative to mobilization of toxic compounds and leaching of nutrients.

o Conducted studies indicating that southeastern watersheds are accumulating sulfur. The importance of this change on the chemical and biological characteristics of surface waters is not clearly understood.

o Completed several studies showing that forest ecosystems alter the acidity of rain traveling through them.

G. Effects on Materials and Cultural Resources:

o Began work to determine possible acid-deposition-related damage to historical monuments.

o Established several field sites for testing materials and estimating the relationship of deterioration rates to acid deposition.

o Began analysis of available materials deterioration data and started documenting performance of possible protective treatments for masonry.

o Tested a simplified deposition monitor for application to material damage research.

H. Control Technologies: (FY 1982 developments under Federal pollution control hardware programs relevant to acid deposition)

o Successfully tested developmental prototype of low-cost, retrofitable technology (LIMB) that can simultaneously decrease sulfur dioxide and nitrogen oxides emissions from utilities by 50 percent (EPA).

o Developed burner systems to decrease nitrogen oxides emissions by 75 percent in large coal-fired boilers (EPA).

o Assembled data on the characteristics of U.S. coals to aid in determining their emission control potential (EPA, DOE).

o Evaluated improved wet and dry sulfur dioxide scrubbing (removal) processes (DOE, TVA, EPA).

I. Assessments and Policy Analysis:

o Prepared the "Critical Assessment Document: The Acidic Deposition Phenomenon and Its Effects" with chapters reviewing acidic deposition and its impacts with contributions from 54 scientists from many institutions in this country and abroad. The document is now undergoing public review and final revision.

o Initiated work on developing methodologies for conducting integrated assessments of the significance of various scientific and economic information.

o Developed the preliminary version of a model for assessing sulfur dioxide emissions from the utility sector.

o Began investigation of estimating uncertainty in models and the implications for formulating control strategies.

J. International Activities:

o Established basis and mechanisms for increased cooperation between the U.S. and Canada on research and monitoring activities.

o Began developing specific proposals for joint research efforts with other nations.

o Published "International Directory of Acid Rain Researchers", listing 1,000 scientists in 32 countries.

More details of these and other accomplishments in the various research categories is contained in the FY 1982 Annual Report of the National Acid Precipitation Assessment Program.

MAJOR FY 1982 PROGRAM DEVELOPMENT ACTIVITIES

In addition to specific research accomplishments, in FY 1982 the Task Force met with representatives of key groups conducting acid deposition research and monitoring activities including private sector and environmental organizations, states, and Canada, to plan and implement increased coordination with these groups. Such activities include:

1. Improving Coordination with Non-Federal Activities. The Task Force is committed to conducting a National Program that coordinates the federal efforts with the research and monitoring activities of the states, private sector groups and other countries, especially Canada. Several specific steps have been taken to develop and encourage such cooperation and more extensive joint planning activities are anticipated.

Besides reviewing the draft National Plan, the Task Force's April 1981 workshop of non-Federal experts began a dialogue with state and private groups.

The workshop report describes the participants' ideas concerning how coordination and cooperation can best be accomplished. The Task Force is implementing many of these suggestions and is actively pursuing continued exchanges of information with non-Federal groups.

At the First Annual Review Meeting of the Task Force (September 1982), approximately 30 non-Federal participants provided valuable ideas to discussions on interrelationships among research areas, using the draft technical Operating Research Plan as background.

Task Force representatives have met with certain non-Federal groups, including private sector, certain states, and Canada who are sponsoring substantial research on acid deposition. These meetings have focused on reviewing programs and establishing a basis for a better understanding and closer coordination. During the last year, the most significant such events were:

U.S.-Canadian Research Program Cooperation

In April 1982 and June 1983, Task Force representatives met with the Canadian Federal/Provincial Research and Monitoring Coordinating Committee (RMCC) to expand bilateral scientific cooperation. Leaders of both the U.S. and Canadian research efforts emphasized the mutual value of greater co-operation and coodination of activities, and many joint efforts are underway.

Federal-State-Private Sector Coordination

The Task Force initiated and encouraged joint sponsorship of an Acid Deposition Research Coordination Workshop in November 1982. The non-Federal sponsors included the Electric Power Research Institute, the American Petroleum Institute, the Gas Research Institute, the Coordinating Research Council, the National Council of the Paper Industry for Air and Stream Improvement. Environmental groups and states were also invited to participate.

The purpose of the workshop was to bring together key Federal and non-Federal managers of acid deposition research to review and discuss the management and coordination of their research planning efforts. Specifically, the two goals of the meeting were (1) to assess the scope of the nationwide acid deposition research effort, and (2) to initiate a continuing process for joint planning and coordination of Federal and non-Federal acid deposition research efforts.

In addition to developing joint statements on research needs and disseminating information, the state representatives agreed to organize by regions and to define their specific areas of expertise. The Task Force agreed to investigate ways to increase opportunities for state and private sector participation in the National Program planning and review groups. Followup activities include further discussion between the Task Force and the private sector and state representatives to work out the details.

National Program Research Reviews

These detailed scientific reviews (1)allow the National Program's research to be reviewed in a comprehensive manner rather than as a series of discrete agency projects; (2)assess the quality of the science; (3)provide a forum for interaction by field level scientists; and (4)evaluate the progress of the various research approaches. The panels of outside experts critically evaluate all research to be sure that each project contributes to the identified objectives, milestones, and deliverables. The review of effects research was held in February, 1983 and the review of atmospheric processes and monitoring research was held in August, 1983. The review of assessment activities is proposed for January, 1984.

2. Developing an Inventory of All Federal- and State-Sponsored Acid Deposition Research. The initial tabulation was completed this spring and the inventory will be maintained and updated for use by the Task Force and all interested parties. This inventory includes information on all ongoing projects with the performer(s), duration, funding, study area, research goals, methodologies, and expected outputs. The private sector has conducted a similar inventory of its activities; together with the Task Force's effort, this should provide a complete catalog of all acid deposition research and monitoring activities in the United States.

OUTLOOK FOR RESEARCH PROGRESS

In the next several years the research under the National Program will produce a number of important results. From FY 1983 to FY 1985 the Task Force agencies expect to:

- Produce a report estimating for different regions, how effective control of sulfur oxides, nitrogen oxides, and/or volatile organic compounds would be in reducing acid deposition.

- Complete assessments of the potential significance of alkaline dusts and natural sulfur and nitrogen emissions in influencing precipitation acidity over sensitive regions.

- Begin applying advanced models to predict utility emissions, and develop a preliminary model to extimate industrial combustion emissions.

- Complete a major transpoet experiment (CAPTEX) to help determine long-range pollutant transport patterns and facilitate evaluation of atmospheric models.

- Develop a model capable of estimating local/mesoscale atmospheric deposition.

- Complete the National Trends Network, to provide adequate coverage to produce reliable annual maps of acid deposition for North America.

- Develop and begin applying improved techniques for monitoring dry deposition.

- Complete inventories and detailed regional maps of the extent of sensitive and currently acidified surface water.

- Prepare a comprehensive report on the status of drinkin water supplies.

- Finish a broad assessment of the potential effects of acid deposition on soils.

- Establish dose-response information on corn and soybeans in eastern and midwestern regions of the United States.

- Complete initial determination of the effect of acid deposition on the susceptibility of forests to drought and other stresses.

- Prepare a report on historical rates of deterioration of building material and complete inventory of materials at risk in the Northeast.

- Complete initial assessment of the scope of material damages caused by acid deposition.

- Finish report on historical trends in emissions, deposition, and effects.

- Develop and apply advanced integrated assessment methods to evaluate the optimal strategies for managing acid deposition and its effects.

While it will take a systemmatic effort over a number of years to adequately address the major uncertainties about the causes, effects, and management of acid deposition, the National Acid Precipitation Assessment Program made an ambitious start and broad progress during its first year of research. The Program has laid the groundwork for continued scientific advancement and the framework has been established to focus the research on answering the questions most critical for developing sound policies. The National Program's recurring assessments will provide a successively refined scientific basis for decision making as well as opportunities to reexamine research directions.

Biographical Sketch

9/1/83

Dr. Chris Bernabo is the Executive Director of the National Acid Precipitation Assessment Program. He serves in this role while on assignment from his position in the Administrator's Office at the National Oceanic and Atmospheric Administration (NOAA).

At NOAA he is responsible for analysis of the agency's research and development programs and has worked on issues such as the CO_2 and ozone problems, the National Climate Program, and weather modification.

Prior to joining NOAA, Chris was a Congressional Science Fellow and served on the staff of the U.S. House Science and Technology Committee.

Bernabo has a Ph.D. in Geological Sciences from Brown University and has conducted research and published papers on climatology, environmental geology and ecology.

Mr. FUQUA. Thank you very much and Dr. Calvert, we are pleased to have you here once again and are interested in hearing your testimony.

STATEMENT OF DR. JACK CALVERT, SENIOR SCIENTIST, ATMOSPHERIC CHEMISTRY AND AERONOMY DIVISION, NATIONAL CENTER FOR ATMOSPHERIC RESEARCH AND CHAIRMAN, COMMITTEE ON ATMOSPHERIC TRANSPORT AND CHEMICAL TRANSFORMATION IN ACID PRECIPITATION

Dr. CALVERT. It's a pleasure to be here——

Mr. FUQUA. You need to pull that——

Dr. CALVERT. Can you hear me?

Mr. FUQUA. I don't think it's on. Maybe you can use the other mike.

Dr. CALVERT. I'll use Chris'; it works.

It's a pleasure to have the opportunity to come and talk to the committee. I appreciate the fact that we've got crises in the world on things that are other than acid rain that take the time of the participants of the various committees and to appear here. Certainly I'll try not to waste your time.

I've been asked, though, by Representative Don Fuqua of Florida and Representative James Scheuer to come and present to you some of the findings of the National Research Council Committee on which I serve as chairman, and I'll try to give this to you very briefly, but I'd like to read pretty much from the statement which most of you have in hand.

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I think it's recognizable from what Chris has presented that there is a multitude of difficulty, a multitude of problems that one has in trying to come up with solutions to acid rain. I don't think anyone's trying to, indeed, invent the wheel and what's being done in Chris' effort. Indeed, he has some problems that he's identified and is working hard on.

In a few minutes, I can't really describe to you the findings of our report, and I don't attempt to do that, and I hope that those of you have time will try to read the report. It, in a sense, was supposed to be written so that the average layman could read it. I doubt that that's been done because of the difficulties that people seem to have had in interpreting some of the statements which end up to be double negatives very often.

But I believe that it is understandable and if you do get a chance, look at that report, you would. It has a pretty cover on it, at least.

The members of our committee, I think, were outstanding gentlemen and outstanding scientists. Let me just mention to you briefly. They seem to represent the gambit of the university and the research laboratories. James Galloway, of course, from the University of Virginia, was on our committee, from Charlottesville; Jeremy Jake Hales, whom I respect highly, from Battelle Pacific Northwest Laboratories, was on our committee; Dr. George Hidy, Environmental Research and Technology, an outstanding contributor, even to the program the Canadians have on their modeling. George's group, as you remember, runs this for them.

We had, as well, Jay Jacobson, from Boyce Thompson at Cornell University in New York; Alan Lazrus—Al is here, by the way, and is the brains back of this report. He is a little man who knows what he's doing and the rest of us are a bunch of theorists. I respect Al's contribution immensely.

John Miller, from the National Oceanic and Atmospheric Administration, from Rockville, Md.; Prof. Volker Mohnen, from the State University of New York, Albany; and our research council staff who was invaluable was Myron Uman.

Now, there is a reason why our committee was established; it wasn't to try to make trouble for the acid rain community, certainly, or to indeed invent some new ideas. We were told to look at the literature and indeed see what is known scientifically and try to make some judgments about what might be done with respect to the relationships between emissions and deposition that would be helpful to people who are trying to make laws and do the difficult things that are built upon judgments that are made scientifically.

It is true that many of our observations that were made were made because we were designated to do them. We weren't told to look over the whole field; we were told to look indeed at one of the problems that was faced by the scientific world that was suggested by the biologists that acid rain deposition should have a 50-percent cutback in order to remove some of the difficulties that were associated with the ecosystems in some of our sensitive areas.

That 50-percent cutback was not something that you could just say we can solve immediately by going and cutting fuel emissions by 50 percent. It was obvious that there was some relationship which need not be linear.

So it was suggested to us, well, tell us what is necessary; how much would you have to cut back in order to accomplish a 50-percent cutback in, say, sulfate deposition, in sensitive areas? So it was this charge which largely led to our study, and I might point out that many of us—and of course, we didn't work for pay, we worked for the interest of the science, indeed, and we worked for about 1½ years doing this in our spare time. I think the people that contributed did their very best job to try to come up with some suggestions which would be helpful, although the final result may not suggest it's a great lot—a great deal of help.

We feel that it's the best we can do at the moment. As Chris has pointed out, the area is very complex and that to try to solve a problem as complex as this with some kind of easy solution is left to the politicians who seem to work magic, but God bless you if you can do so. This is a very difficult area.

Let me summarize very briefly some of our findings. First, and this is in agreement with what Chris has said and certainly Bill will say in a moment if he reads from his sheet that he has there. We do feel that it's important to recognize that there are naturally occurring acidic substances, that it's just as important to identify that in northeastern United States, there is no way we can explain the acidity by these natural occurring acidic substances.

I think one has to identify with the fact that, indeed, we, the polluters of America, have contributed to the problems that exist in the Northeast, and indeed, without taking any hesitation, I would say that all of us have contributed to the acid deposition in the Northeast, and all of us are to blame for it.

We all use energy and I think in hot, humid Wasington, D.C., I can see why you would want to use air-conditioning in the summertime. We in Colorado don't need to do that sort of thing; we can conserve our fuels, but I think we do all have to take a responsibility for the fact that about 90 to 95 percent of the acid deposition arises from manmade emissions.

We're talking about eastern—northeastern North America, now, when we're talking about that. There are many areas of the United States that aren't complicated in that fashion.

The complex chemical/physical process which occur both in the atmosphere and in clouds transform the primary emissions, namely the sulfur oxides and the nitrogen oxides, into the major acidic products which we have observed. These, of course, are sulfuric acid, nitric acid, and the ions that are derived from those acids.

Important for our consideration is the manner in which these complex chemical and physical processes relate to acid deposition and emissions. If we are to control emissions to achieve a desired acid deposition level, relationships, of course, that relate emissions to depositions would prove very valuable. It isn't a trivial problem. If you read from my script here, at least I make the case that indeed we should not expect this to be a linear or proportionate operation between emissions and deposition. All of the committee came in with this expectation our guesses were from 10 to 40 percent reduction would occur if one managed to cut back by 50 percent on emissions.

The tune changed somewhat as we, indeed, studied the literature and saw the evidence. Our suggestion, though, was to not go into

this expecting linearity, but to expect nonlinearity, or disproportionate response.

This is why our conclusions are based in the negative, double negative sense. We're talking about there not being strong disproportionate or nonlinear effects. That's done because everyone expects it to be nonlinear. So the observations that we have are phrased in that double negative sense not to be mean but because we are forced to by the times.

If we move to really what our conclusions are, which I think you would really like to hear, instead of my rambling here about the importance of our study, our charge was to look at what the relationships were and these relationships are not clear and our suggestions are based on our best expectations indeed.

By the end of our deliberations, though, our committee was unanimous in its agreement of the report's findings and conclusions, and let me just briefly go over those with you.

First, for yearly averaged emissions and depositions over large areas of eastern North America, there's no evidence for a strong disproportionate or nonlinear relationship between emissions and depositions. When we are talking about linearity now, we mean that a 50-percent cutback in emissions would result in a 50-percent cutback in depositions from that particular emitting source, integrating over the area that this source affected.

Or a 10-percent cutback, affected a 10-percent cutback in depositions. That's what we mean by now linear. We are not trying to be, indeed, scientific or nonscientific in the use of that word. Since it has been used extensively, we've sort of stuck with it.

In trying to come up with our conclusions, we've studied available data, and as Chris has pointed out, that available data in the United States is rather meager. The historic trends in emissions and depositions in New Hampshire, about the only one that we could reliably use—oh, there are data that certainly have been collected in many of the agencies, and we do not mean to do them in by saying that these are the only reliable set; they are the only ones that have self-consistent use of analytical techniques and multiple samples run so that one has confidence in what one finds.

The historical record at Hubbard Brook, at least that matched what we had available to us in the way of emissions extended over 13 years, and it indicated to us that a deposition in that area had tracked changes in emissions in the Northeast. Our conclusions are consistent with expectations that are based upon current theory. It isn't magical that this would happen, indeed.

The—they are certainly consistent with expectations based on current theory related to the mechanism of acid generation provided that the oxidant deficiency is unimportant and rainwater samples, which deposit acids in sensitive areas in Eastern United States during the active periods for acid deposition; namely, the summer months. We could include there the spring, summer, and the fall months; summer being the most active in depositing acid rain.

Certainly preliminary observations which we have during the past year from White Face Mountain that Dr. Lazrus and his group have done in conjunction with Dr. Mohnen suggest that this hypothesis is correct, that there seems to be, indeed, hydrogen perox-

ide in excess of the sulfur dioxide present in those areas, and if that is true—and we don't know that's true irreversibly in the summertime, but if that were true in the United States, one would expect in theory a proportional relationship between emissions and deposition.

Indirect evidence based on the patterns of the ratio of sulphate ions to nitrate ions in deposition also did not support the evidence of a strong disproportionate or nonlinear relationship between sulfur dioxide emissions and sulphate deposition in eastern North America.

The ratio is relatively constant throughout the area of eastern North America in which the acid precipitation is high. These ratios resemble the ratios of sulfur dioxide and nitrogen oxide emissions averaged on an annual basis by State or province.

Whatever the nature of the complex chemical and physical processes which occur in acid deposition, the surprising result is that if you average over a year, there's a near equal fraction of both sulfur and nitrogen oxides which are oxidized acids and deposited in precipitation in the Northeast.

These considerations suggest that on an annual average basis, there's no evidence of a disproportionate relationship between emissions of acid deposition over a wide area of eastern North America.

All right. I've used the phrase "no strong disproportionate or nonlinear relationship." I've explained somewhat what I meant by that, but strictly speaking, any relationship between emissions and depositions is either proportionate or it's disproportionate.

You have to say yes or no in law and you have to say it certainly in science eventually, too. It's highly unlikely that any strictly proportionate or linear relationship exists in this case as well. However, the important consideration that we have to face is how significant are the disproportionate or nonlinear processes on the average.

We concluded that within the uncertainties associated with the errors in the data and in estimating emissions, disproportionate or nonlinear processes are apparently unimportant for annual averages over large areas of eastern North America.

We feel that it's a reasonable hypothesis that the relationship is essentially linear for long-term averages over large areas. Of course, this doesn't say anything about the degree of proportionality or linearity between emissions from a given source and deposition of acids at a given site during a specific long-range transport event. Certainly theory would suggest that there would be times when this would be very nonlinear.

Let us turn to the second issue now: What is the relative importance of long-range transport from distant sources compared to nearby sources? The question is an important one, as you recognize, because we have sensitive areas that have powerplants. Oswego, N.Y., for example, spewing out a lot of emissions right near the Adirondacks; Eastman Kodak Co. with a power emission that is not particularly well controlled. All these contribute, of course, to the Adirondacks. The extent to which they contribute now is something which is unknown to me, and certainly it is something that is unknown to science.

It is very difficult to say how those contribute compared to, say, the emissions that are in the—my favorite hometown, Columbus, Ohio, or on the Ohio River. These certainly contribute. Everything in the United States contributes to one extent or another. The question is, what contributes what.

And I think this is the problem that the scientists face today, trying to say what does what? We don't want to control things we don't have to control. If Ohio is willing to live with their high sulphate and their high nitrate, let them do so, if they aren't contaminating the rest of the world.

But the fact is we don't know right now very good ways to do evaluation of the source apportionment.

All right, anyway, let me mention to you what our thoughts are on this subject. There is good theoretical evidence that long-range transport occurs and I don't think there is any doubt about this. It's common sense that we suggest that if there were two equivalent sources upwind from a sensitive area, the closer one was likely to have the greater impact.

I think I've have been criticized by my friends who are good modelers who say, "Well, hell, Jack, you can say something better than that. We know what we're doing when we model these things."

Indeed, we do, but very often, the modeling that's been done with Gaussian plumes—if you try to extrapolate this to long-range transport, it's like predicting the weather, you know, some of the time you're right, some of the time you're not right. And that's the kind of thing I think one has to face in making policy judgments, and indeed we don't build a strawman that somehow science is expected to produce for us some miracle and the fact is that I would doubt that anybody has a model today he could trust as well as a weather map to, you know, give you the final answer.

There's a probability it's right; there's a probability it's wrong.

All right. If we talk in terms of what we're saying; we say that we cannot now reliably determine the relationship between emission from a specific source and deposition and a specific receiving area. That is, we don't know specific source receptor relationships. Long-range transport models which are now in use do not incorporate the detailed chemistry or the physics which are necessary to define well the rates of acid generation and transport processes which occur in the atmosphere in the clouds.

This is the not the fault of the modelers; they don't—they haven't really got a lot of field data they can build a model on. The field data that have been determined over the United States has been very restricted, aside from some sure data and some other studies that have been very important in formulating models, we haven't really any long-term record that can tell us what the model will predict and what success that model will have.

Based on the study of acid deposition phenomena, we suggest strongly that additional studies are needed to provide the data required to develop more accurate models that can assess properly what's doing what.

Now in our opinion, studies of both physical and chemical processes that are involved in the actual motions and the transformations in air parcels under meterological conditions that are typical

of acid rain episodes in Eastern North America are especially important.

Who cares what's happening in somebody's laboratory chamber? Really, what's happening in the real world is what we ought to be concerned with and it seems to me field studies dedicated to looking at acid rain and how it's made as it's being made are going to be the most valuable in doing this.

I might—apply our findings to the analysis of alternate control strategies. This is where I feel that we've probably given you a kick in the pants rather than a lot of help, but let me indeed suggest to you what we feel our report does support at least.

If sulfur dioxide emissions were changed throughout eastern North America in such a way that the spacial pattern of emissions remained unchanged, then we'd expect that a similar change in the deposition of sulfate—derived from sulphuric acid—would occur. The result also applies only to changes in emissions of sulfur dioxide, those of other pollutants, including the nitrogen oxides and hydrocarbons remaining unchanged. If sulfur emissions were substantially reduced in the aggregate but reductions occurred only in specific geographic regions, for example, in areas where emissions currently are the highest, we'd expect total deposition over Eastern North America to be similarly reduced, but we can't predict that substantial reductions would occur specifically in the areas threatened by acid rain.

I think even the best models that we have today, and certainly there are good modelers; certainly Jack Shannon at Argonne, some very important and very successful modeling that's been done by their group has shown that, indeed, you can make predictions about what's doing what, and you have in your hands, I'm sure, reports that suggest what's doing what. Suggestion of the current models are maybe that 20 percent of the sulfate deposition in the Adirondacks at most would be coming from that midwest region, maybe even less, 14, 15 percent some models suggest.

We can't just indeed cause a reduction of the acid precipitation in the Northeast by causing Ohio to give up developing power. If we had them stop developing power completely, the effect would be to cut back emissions by 15 percent in the Northeast. That's a big help. Ohio should help us do that, indeed, since they're the biggest polluters in the world, probably, but the fact is that you can't have one man's participation and expect a result. I think the thing we need is a cooperative effort that sort of overcomes the difficulties we have as politicians, I'm sure, to do things that are for the—not only the good of the party, but for the good of the whole country—that we do work for some kind of common agreement. Look, we've got to do something about acid rain and there are some things we can do and they all don't depend on Ohio. They depend on New York and Pennsylvania and the Midwest and probably elsewhere.

As soon as we buy the fact we're all contributors to this, I think there'll be less bickering and less difficulty, perhaps, in finding an economic base that probably can solve the problem.

All right, if we wish to apply the operations of computer strategy to the techniques that are outlined to us by control agencies these days, let's do so with a jaundiced look at what's being done, indeed, and let's ask the man who's doing this to provide us with the error

limits. What is the chance that he's right? He's like a weather man, he ought to tell us what the probability is. If there's going to be this effect or that effect.

Let me get off my soapbox and respond now to what you've asked me to respond to as well. There is a acid precipitation assessment program that I respect very highly that Chris has described to you. I think the education that he's given us all with the slide show is certainly more than any education I can give you with some words.

He has expressed, I think, the problem rather succinctly. I consider that annual report of the President—to the President and Congress of the national acid precipitation assessment program to be a valuable contribution. You recall that the National Laboratories were given this 10-year mandate by Congress for this effort under the Acid Precipitation Act of 1980.

My feeling is that the direct involvement of scientific staff of the National Laboratories to the solution of a major national problem is a most commendable goal and something that I wonder why we don't do more of. The National Laboratories are highly motivated, good scientists.

The multipronged attack of the problem in which field studies and laboratory studies and modeling efforts all interact closely in the search for solutions is the most reasonable plan. There's no question in my mind that useful information can be obtained in the efforts of the groups which planned this document.

Quality in many of the scientists which are associated with the program of the National Laboratories is good and their careful thought in the preparation of the document is certainly evident.

The state of the science as outlined by the task force is reasonably accurately summarized, I feel, and it is in general accord really with the recent report of my NRC committee. For example, it is pointed out that manmade atmospheric pollutants are probably the major contributors of acid deposition in Northeastern—Northeastern North America; that natural sources are believed to be minor contributors to acid deposition; the task force concludes, and I concur, that the ability of current generation models to predict source receptor relationships with adequate resolution accuracy is an unresolved issue.

Research activities related to natural sources, manmade sources, atmospheric processes, deposition monitoring, aquatic effects, and terrestial effects; they represent the entire gamut of the important areas of research related to acid deposition. The specific research effort's not outlined in sufficient detail from this document for me to assess critically what the proposed programs really are doing in detail, but as Chris has pointed out, there is an indepth review underway now. There is a—there has been a weeklong session in Boston, which was indeed attended by presumed experts, namely, academicians and people in research-oriented programs throughout the world that came in to evaluate this program.

Remember that under the President's Council on Environmental Quality, there's also Prof. John Deutsch of MIT that's been appointed to, indeed, evaluate this program, so I'm sure, with the careful reviews that the project is getting, if there are any problems there, if they're not discovered and pointed out to Chris, he's got—he's indeed missing something, as I'm sure he'll have some

helpful input from all the whizzes of the world that they bring from the academic community, like myself.

In the opinion of this reviewer, and let me point out that my comments on these reports, Chris, are not those of my committee because there wasn't time to really circulate that to the entire committee. These are my opinions and you can weight them particularly low if you wish to, Chris, but in my opinion—this is mine—it's unfortunate the plans for the acid deposition research focus entirely on the efforts of the national laboratories. Chris has pointed out that there is this NSF fund that supports academicians; if you ever applied for an NSF grant, you realize that's really not much in support; it's pretty primeval.

There is a large untapped resource of very great value, the university research community, which I think could contribute uniquely to a national effort, and it seems to me that as you legislators plan the future of this, I would—I'm not in the academic committee so I'm not grinding an ax for myself. I feel this would be an important asset to the program.

From my perspective with a rather limited knowledge of the details of the entire national program, I would conclude that the participating laboratories appear to be addressing the right questions; they are planning studies to obtain answers which should be very important to providing a better scientific base for the description of acid deposition processes.

Let me turn my attention now just in closing here to one other report that has appeared, and Bill's going to tell you about this report, and it's unfair for me to comment in advance of that, but remember that Presidential Science Adviser, George Keyworth, the Office of Science and Technology appointed what they call an acid rain review panel and this panel has come up with—at least as a preliminary report that we received a message from not too long ago. Remember that this committee did review the United States-Canadian scientific studies and they formulated several important conclusions that bear on the problem.

In general, I concur with their observations. In my opinion—now, Bill, this is my opinion, again, not my committee's—but they did conclude, as did our NRC Committee, that acid deposition has no acceptable method now for determining source receptor relations. The community is stuck with the fact that on a scale smaller than eastern North America, it is very difficult to indeed attribute given sources to given receptor areas.

They suggest that many years—possibly 10, but certainly not 5 is the way they phrased this—I think that's sort of a dig on us because we used the word "five," of research effort would be required to understand the diverse biological, chemical and physical aspects of the problem.

Our NRC Committee on Acid Deposition was also pessimistic about the time required for the complete quantitative description of the chemical and physical processes which govern the acid development and deposition. We were more optimistic about the time scale for the determination of useful information related to the delivery of acids to rural areas by transport and transformation processes through direct empirical observations in the field. We feel that this might provide the basic phenomenological evidence with

sufficient reliability to form the basis for improving the near-term strategy for dealing with acid deposition problems in Eastern North America.

I endorse the conclusion expressed by the acid rain peer review panel that acid deposition constitutes a problem for which solutions should be sought; that reduction in the emissions of sulfur compound in the atmosphere should be made beginning with those steps which are most cost-effective.

I am not qualified to evaluate the concerns of the committee related to the potential biological impact of acid deposition, and in particular, the potential for irreversible damage to denitrifying microbes in acidified soil, however, it is obvious that the quality of this science who are on this committee, the scientists are exceptionally highly qualified and we would be well advised to consider their comments very carefully.

I look forward to the release of that complete report which I'm sure you're busy working on in your spare time.

Let me mention one last thing and this last effort is one that I hope you don't take as a personal attack on any individual, any person, but since there is widely circulated a review of our NRC deposition report, I feel obligated to at least stand up for my committee and indicate that probably it's wise that we don't use the Halls of Congress to argue important scientific issues. I don't intend to do that.

A congressional hearing or a Congressional Record isn't well suited to scientific debate, but there was a report that was released—namely under the guidance of Bernie Manowitz of Brookhaven National Laboratories—and it presents to you the selected representatives of three national laboratories supported by the Department of Energy; namely, Brookhaven, Battelle Pacific Northwest, and Argonne.

These reviews were sent to the National Academy—my bosses—you can imagine my embarrassment when I just got through with this report and here appears a fairly critical report of our report, not in the scientific literature, but in a report to our—my peers at the National Academy, but to various members of the interagency task force, Chris and his colleagues. My friends in industry and Government tell me the report was widely circulated to the leadership and research arms of the power industry, the related industrial operations throughout United States and within the extensive communications pipeline of the administrative and legislative staffs of the Federal and State governments.

The original report, I think you don't have in hand, many of you don't, maybe. Many of you do. The original report was subsequently sanitized and presented to Representatives Fuqua, Scheuer and Lloyd with additional comments from a group at Oak Ridge National Laboratory, and this has been, or will be introduced in the Congressional Record.

We didn't have access to this revised version of the reviews and I won't spend a lot of time on it, but because of that fact, I can't respond to that most recently received one, although scientific questions remain unchanged.

I've submitted as appendix D to the formal report of the Congressional Record the original Manowitz group review, which should be

published as part of the record of this hearing. Comments given here represent those of only two of our committee, namely myself and Al Lazrus, whom you've met, since the time didn't allow participation of our entire committee.

All right—what——

Mr. SCHEUER [presiding]. Dr. Calvert.

Dr. CALVERT. Yes.

Mr. SCHEUER. You see we have an enormous number of members here today, and we're not going to have any time left for questions for the members, so we're going to ask you to summarize the rest——

Dr. CALVERT. All right.

Mr. SCHEUER [continuing]. Of your testimony so we can get on to Dr. Ackermann and then have some time left for the members to ask questions.

Dr. CALVERT. All right, I will do that. You have access to what I've written and you can take it for what it's worth.

Mr. SCHEUER. Yes.

Dr. CALVERT. I don't intend it to be a boring exposé.

Mr. SCHEUER. I apologize because we've been a little bit lax this morning, but several members have just walked in and we have about eight members, and so we have to be out of here by 12 o'clock, so——

Dr. CALVERT. I understand.

Mr. SCHEUER. OK.

Dr. CALVERT. Let me summarize if I may.

One of the things that I'd like to put across to you that we're not against models of the acid deposition process. There are definitive uses that are made of models and we continue to use them.

Our caution relates to their inappropriate use. If long-range modeling involving the attribution of specific sources to a specific receptor site is used today--and of course, it is used today—then modelers should provide the user with a realistic estimate of the magnitude of the uncertainty associated with the results so that these can be incorporated in the control planning.

Let me just summarize by saying that such a practice is not suggested to frustrate the control plan; that's not our intention at all. It is necessary to provide the sensor realism that is demanded in the formulation of sulfur dioxide emission control strategy in view of the very high costs that are associated with it.

Certainly the conclusions of the NRC Committee on Acid Deposition are necessarily very limited in nature, and indeed, they must and will be checked by extensive further experimentation. Our conclusions represent the best judgment of a group of highly qualified and diversified scientists and we certainly would not alter or compromise these conclusions on the basis of any unusual review of the Manowitz group.

Thank you, sir.

[The prepared statement of Dr. Calvert follows:]

ORAL PRESENTATION AND PRESS RELEASE

REPORT OF THE FINDINGS OF THE NATIONAL RESEARCH COUNCIL COMMITTEE ON ATMOSPHERIC TRANSPORT AND CHEMICAL TRANSFORMATION IN ACID PRECIPITATION PRESENTED TO THE SUBCOMMITTEES OF THE HOUSE COMMITTEE ON SCIENCE AND TECHNOLOGY, SPETEMBER 13, 1983 BY JACK G. CALVERT, NRC COMMITTEE CHAIRMAN

It is a pleasure to appear before the Committee on Science and Technology of the United States House of Representatives at the request of Representative Don Fuqua of Florida, Chairman of the Subcommittee on Energy Development and Applications and Representative James Scheuer of the state of New York, Chairman of the Subcommittee on Natural Resources, Agriculture Research and Environment. They have asked me to present the findings of the National Research Council Committee on which I served as Chairman and to respond to findings of two other recent reports and one informal "review". The report of our committee, "Acid Deposition: Atmospheric Processes in Eastern North America", was published and released recently by the National Academy Press.

One cannot in a few minutes time present all of our findings and the reasoning which our report contains. I do hope that those directly involved in the research, control strategy development, or the legislative efforts related to acid deposition will refer to the complete report for a more detailed and meaningful presentation of our work. First, let me tell you about the membership of our committee. They were appointed according to the procedures of the National Research Council. They are all active research scientists with both national and international reputations for their outstanding contributions to the various scientific disciplines which bear heavily on the acid deposition problem. They were drawn from scientists of the academic community, government and national laboratories,

and private research companies. In my opinion, a more competent and widely representative committee would be difficult to assemble. The members of our committee were Professor James Galloway of the University of Virginia, Charlottesville; Dr. Jeremy Jake Hales of the Battelle Pacific Northwest Laboratories, Richland, Washington; Dr. George Hidy of the Environmental Research & Technology, Westlake Village, California; Dr. Jay Jacobson of the Boyce Thompson Institute, Cornell University, Ithica, New York; Alan Lazrus of the National Center for Atmospheric Research, Boulder Colorado; Dr. John Miller of the National Oceanic and Atmospheric Administration, Rockville, Maryland; and Professor Volker Mohnen of the State University of New York, Albany, New York. Our National Research Council Staff Officer was Dr. Myron F. Uman.

Let me describe briefly the reason that our committee was established and the motivation for our report. In late 1981, the National Research Council issued a report titled, "Atmospheric Biospheric Interactions: Toward a Better Understanding of the Consequences of Fossil Fuel Combustion". The main focus of that report was on the effects on living systems of atmospheric pollutants associated with energy production. In one chapter which dealt with the effects of acid precipitation on the biosphere, the committee identified the maximum upper levels of acidic deposition which could be tolerated by sensitive lake ecosystems; this level is the lowest acidity at which rates of degradation are detectable by current survey methods. In several specific areas of the United States and Canada, where neutralization of deposited acids by the soils is not efficient, acidities have been found that are higher than those identified as potentially harmful to lakes. The committee at that time suggested that a cutback of acid deposition in these sensitive areas by

50% would be necessary to avoid adverse effects. To implement this or any other goal of reduced acid deposition, we are faced with the obvious question, "How much reduction in emissions of the acid precursors will be necessary to achieve the goal?"

The search for the answer to this question, not in the charge of the former committee nor addressed by them, was designated as the major task of our committee. The largest part of our effort involved a thorough review of the scientific literature which describes the theory, experiments, and field studies and assessment of the current scientific understanding of the atmospheric processes involved in the phenomenon of acid deposition in eastern North America. There is no need to discuss here all aspects of this review. Let me summarize briefly a few of the most important observations.

First we recognized that naturally occurring acidic substances and basic substances which enter the atmosphere can influence acid deposition. However, in most areas of the northeastern United States there is a high probability that a very large share of the acidity of atmospheric deposition (about 90-95%) arises from man-made emissions, largely sulfur and nitrogen oxides. Complex chemical and physical processes which occur both in the atmosphere and in clouds transform these primary emissions into the major acidic products which we observe, sulfuric acid, nitric acid, and the ions derived from these acids (hydrogen ion, sulfate ion, and nitrate ion). Important for our considerations here is the manner in which these complex chemical and physical processes relate the acid deposition to the emissions which are released. If we are to control emissions to achieve a desired acid deposition level, such relationships would prove to be very valuable.

At first consideration one might feel that the problem is trivial. Why wouldn't a specified reduction in the emission of oxides of sulfur and nitrogen result in a proportional decrease in acid deposition in sensitive areas?

That is, why wouldn't a 50% reduction in emissions result in a 50% reduction in acid deposition, or a 10% reduction in emissions result in a 10% reduction in acid deposition? There are many reasons why such a result cannot be expected at first sight. First, nonlinear (or disproportionate) responses to changes imposed on natural systems are most commonplace. An important example of this is the case of the generation of ozone and other oxidants in polluted urban atmospheres. It is well established that a given percentage reduction in emissions of oxides of nitrogen and hydrocarbons, the most important precursors of urban smog, leads to much less than a proportional reduction in ozone generated in the polluted atmosphere. After many decades of research by scientists, theory and experiments have detailed the reasons for this complex disproportionate (nonlinear) character of the ozone-nitrogen oxide-hydrocarbon system.

Similarly there are disproportionate or nonlinear interactions between the chemistry of the sulfur dioxide and that of nitrogen oxides in the atmosphere which must be considered. All of the various pathways that lead to the transformation of sulfur and nitrogen oxides to acids are coupled by common products and reactants that can directly and indirectly influence the rates of reactions by other pathways. For example, it has been hypothesized that the competition between sulfur and nitrogen oxides for oxidizing species in the atmosphere, as well as oxidant limitation, suppression of oxidation because of acid build-up and other processes may lead to disproportionate or nonlinear relations between emission cutbacks and observed cutbacks in acid deposition. In the absence of definitive evidence to the contrary, therefore, we would be ill-advised to assume that a directly proportionate relationship will necessarily exist between the response of acid deposition in sensitive areas and reductions in the emissions of acid precursors.

Of course we recognize that the establishment of source-receptor relationships is a very important task of science in that the economic factors related to control are very dependent on this relationship. If the relationship is nearly proportionate (linear), the costs of emission controls to achieve a desired reduction in acid deposition in a given region are much less than those required to achieve the same degree of reduction in deposition in this region if the response is very disproportionate (nonlinear). The establishment of source-receptor relationships is important in other ways as well. If, for example, sources in a certain geographic area can be shown to have the predominant effect on a specific ecologically sensitive receptor area, a detailed control strategy might be designed to protect that area at minimum expense through the control of only a few sources. If such relationships cannot be demonstrated, then we may be left with more widely applied control strategies which may be much more expensive in the aggregate.

In addition to our study of the theoretical aspects of the chemical and physical phenomena as they relate to source-receptor relationships, a great deal of our study dealt with empirical observations of acid deposition within the eastern North America. Although these observations are very limited in number today, they show directly the ultimate results of the occurrence of all the incompletely understood, complex chemical and physical processes which result ultimately in acid generation, transport and deposition.

By the end of our deliberations, the committee was unanimous in its agreement with the report's findings and conclusions which I present to you now. First, for yearly averaged emissions and deposition over a large area of eastern North America, there is no evidence for a strong disproportionate (nonlinear) relationship between emissions and deposition. This finding is based on several

scientific considerations. We analyzed the available data on historic trends in emissions and in deposition at one location in New Hampshire (the only reasonably reliable historical record in North America), and on the ratio of amounts of sulfur dioxide and oxides of nitrogen in emissions and the ratio of the products deposited as sulfate and nitrate in precipitation. The historical record (over 13 years) indicates that deposition has tracked changes in emissions. Our conclusions are consistent with expectations based upon current theory related to the mechanism of acid generation provided that oxidant deficiency is unimportant in rain water samples which deposit acids in sensitive areas of eastern United States during the most active period for acid deposition, the summer months. Preliminary observations during the past year on White Face Mountain in the Aderondacks support this hypothesis.

Indirect evidence based upon patterns of the ratio of sulfate ion to nitrate ion concentrations in annual deposition also do not support the existence of a strongly disproportionate (nonlinear) relationship between sulfur dioxide emissions and sulfate deposition in eastern North America. This ratio is relatively constant throughout the area of eastern North America in which the acid precipitation is high. These ratios resemble the molar ratios of sulfur dioxide to nitrogen oxide emissions averaged on an annual basis by state or province. Whatever the nature of the complex chemical and physical processes which occur in the acid deposition phenomenon, a near equal fraction of both sulfur and nitrogen oxides are oxidized to acids and deposited in the precipitation over the period of a year.

These considerations suggest that on an annual average basis there is no evidence of a disproportionate relation between emissions and acid deposition over a wide area of eastern North America. They show that for the yearly averaged data over a large area of eastern North America the net oxidation of

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sulfur dioxide leading to sulfate in precipitation is as efficient as the net oxidation of oxides of nitrogen leading to nitrate in precipitation in this region. No inhibition of sulfate generation by oxides of nitrogen is evident in the yearly averaged data over large areas of eastern North America.

I have used the phrase "no evidence of a strong disproportionate (or nonlinear) relationship." What does this mean? Strickly speaking, the relationship between emissions and deposition is either proportionate (linear) or disproportionate (nonlinear). In fact, as I have indicated, many atmospheric processes are disproportionate (nonlinear). It is highly unlikely that a strictly proportionate or linear relationship exists in this case as well. However the important consideration is how significant are the disproportionate (nonlinear) processes on the average? Within the uncertainties associated with errors in the data and estimating emissions, we conclude that the disproportionate (nonlinear) processes apparently are unimportant for annual averages over large areas of eastern North America. We feel that it is a reasonable hypothesis that the relationship is essentially linear for long term averages over large areas. Of course this says nothing about the degree of proportionality (linearity) between the emissions from a given source and deposition of acids at a given site during a specific long range transport event.

Now let us turn to the second issue before us: What is the relative importance of long-range transport from distant sources compared with nearby or local sources? The answer to this question is not trivial either. Theoretical and observational evidence exists for the long-distance transport of air pollution leading to acid deposition. There is no doubt about this. Of course, common sense suggests that of two equivalent sources upwind from a sensitive area, the closer source is likely to have the greater impact. However, in realistic cases we don't have equivalent sources close to and far from the areas which have been identified as needing protection. Emissions in the vicinity of ecologically

sensitive areas are generally considerably less than emissions from more distant and more highly industrialized areas. Based on our analyses of both available data and currently available computer models of long-range transport, we conclude that the relative importance of long-range and nearby sources to the acidification of sensitive areas cannot be determined with reasonable certainty at this time. We also cannot determine reliably the relationship between emissions from a specific source and deposition in a specific receiving area. That is, we do not know specific source-receptor relationships. Long-range transport models which are now in use do not incorporate the detailed chemistry or the physics which are necessary to define well the rates of acid generation and the transport processes which occur in the atmosphere and the clouds. Furthermore field data are too limited at this time to develop empirical models which can be validated over the range of conditions which are sufficiently broad for control policy development. The success of control strategies built upon current unsophisticated models of long-range transport and acid deposition is unpredictable at best.

Based upon our study of the acid deposition phenomenon we suggest strongly that additional field studies are needed to provide the data required to develop more accurate models. In our opinion studies of both the physical and chemical processes involved in the motions and transformation in air parcels under meteorological conditions typical of acid rain episodes in eastern North America are especially important.

How might our findings be applied to the analysis of alternative control strategies? If sulfur dioxide emissions were changed throughout eastern North America is such a way that the spatial pattern of emissions remained unchanged, then we would expect that a similar change in the deposition of sulfate (derived from sulfuric acid) would occur. The result also applies only to changes in

emissions of sulfur dioxide, those of all other pollutants including the nitrogen oxides and hydrocarbons, remaining unchanged. If sulfur emissions were substantially reduced in the aggregate but reductions occurred only in specific geographic regions (for example, in areas where emissions currently are the highest), we would expect total deposition over eastern North America to be similarly reduced, but we cannot predict that substantial reductions would occur specifically in the areas threatened by "acid rain". That is, using the currently available scientific data, we cannot specify which sources are responsible for deposition at particular ecologically sensitive sites; therefore we cannot analyze the consequences of control strategies that would require sources in different regions to reduce emissions by different percentages. However we do not wish to imply by these conclusions that a policy of uniform emission reduction is the best one to adopt. Obviously any control policy decision should be based upon sound economic and benefit analysis of alternative control-strategies. Such an analysis was specifically excluded from our charge, and our committee is not qualified to speak with authority on this aspect of the problem.

APPENDIX A: RESPONSE OF JACK G. CALVERT TO THE ANNUAL REPORT OF THE NATIONAL ACID PRECIPITATION ASSESSMENT PROGRAM TO THE PRESIDENT AND CONGRESS

I have been asked by the Subcommittee Chairmen, the honorable Don Fuqua and James Scheuer, to give my response to the two other major acid rain reports which have appeared recently. I am happy to do this. However it should be recognized that this is my response alone and not that of my committee as there was not enough time to contact the committee membership and receive their input.

First consider the Annual Report (1982) to the President and Congress of the the National Acid Precipitation Assessment Program which was prepared by the Interagency Task Force on Acid Precipitation. You will recall that the national laboratories were given a 10 year mandate by Congress for a research effort related to acid deposition under the Acid Precipitation Act of 1980. The direct involvement of the scientific staff of the national laboratories in the solution to a major national problem is a most commendable goal. The multi-pronged attack on the problem in which field studies, laboratory studies and modeling efforts all interact closely in the search for solutions is a most reasonable plan. There is no question in my mind that much useful information can be obtained in the efforts of the groups which planned this document. The quality of many of the scientists which are associated with this program in the national laboratories is good, and their careful thought in the preparation of this research plan is evident.

The state of science as outlined by the Task Force is reasonably accurately summarized, and I feel that it is in general accord with the recent report of my NRC committee on acid deposition. It is pointed out that man-made atmospheric pollutants are probably the major contributors to acid deposition in northeastern North America. Natural sources are believed to be minor contributors to acid

deposition. The Task Force concludes, and I concur, that the ability of current generation models to predict source/receptor relationships with adequate resolution and accuracy is an unresolved issue.

The major research questions which the Task Force has recognized as important for resolution are: (1) the biological issues related to acid deposition; (2) identification of the sensitive resources, the damage which may result, and the timing of these effects; (3) the extent of wet and dry deposition of acids; (4) the relative importance of local versus distant pollution sources in controlling the acidity of deposition at a given location; (5) the relative contribution of man-made versus natural sources of acid-forming material; (6) the temporal and spatial distribution of acid deposition; (7) what changes in deposition would result from a given change in sulfur dioxide, nitrogen oxides, organics, and/or primary sulfate emissions. In consideration of the latter problem they concluded that scientists cannot precisely calculate the amount of emission reduction that would be required to reduce acid deposition below a particular level within a given area, a conclusion similar to that made by the NRC commitee on acid deposition.

The research activities related to natural sources, man-made sources, atmospheric processes, deposition monitoring, aquatic effects, and terrestial effects represent the entire gambit of important areas of research related to acid deposition. The specific research efforts are not outlined in sufficient detail in this document to allow a critical assessment of the proposed programs. However an in-depth review is underway now; there was a week-long review of the acid precipitation programs supported by the federal government held a few weeks ago in Boston, and the planned review of the President's Council on Environmental Quality under Professor John Deutsch of the Massachussets Institute of Technology involves many outstanding scientists who, I am sure, will provide the careful review of the many projects now under study or planned by the national laboratories. In my opinion it is unfortunate that the plans for acid deposition research focus entirely on the efforts of the national laboratories. A large untapped resource of very great value, the University research community, could contribute uniquely to this national effort as well.

From my perspective with the rather limited knowledge of the details of the entire national program, I conclude that the participating laboratories appear to be addressing the right questions and planning studies to obtain answers which should be very important in providing a better scientific base for the description of the acid deposition processes.

APPENDIX B: RESPONSE OF JACK G. CALVERT TO THE PRELIMINARY REPORT OF THE ACID RAIN PEER REVIEW PANEL APPOINTED BY THE PRESIDENTIAL SCIENCE ADVISOR, DR. GEORGE KEYWORTH

Following the requests of Representatives Fuqua and Scheuer, I will comment briefly on the interim report from the Acid Rain Review Panel appointed by the Presidential Science Advisor, George Keyworth of the Office for Science and Technology Policy. Of course I have not yet seen the final report of this panel, so my comments necessarily refer to the brief press release only. From their review of the United States-Canadian scientific studies they formulated several important conclusions which bear upon the acid deposition problem. In general I concur with their observations. They conclude as did our NRC committee on acid deposition that there exists now no acceptable method for the determination of source/receptor relationships on a scale smaller than eastern North America. They suggest that many years (possibly ten, but certainly not five) of research effort will be required to understand the diverse biological, chemical, and physical aspects of the problem. Our NRC committee on acid deposition was also pessimistic about the time required for the complete quantitative description of the chemical and physical processes which govern the acid development and deposition. We were more optimistic about the time scale for the determination of useful information related to the delivery of acids to rural areas by transport and transformation processes through direct empirical observations in the field. We feel this could provide the basic phenomenological evidence with sufficient reliability to form the basis for improving the near-term strategy for dealing with acid deposition problems in eastern North America.

I endorse the conclusion expressed by the Acid Rain Peer Review Panel that acid deposition constitutes a problem for which solutions should be sought and that reduction in emissions of sulfur compounds into the atmosphere should be made, beginning with those steps which are most cost effective. I am not qualified to evaluate the concerns of the committee related to the potential biological impact of acid deposition, in particular, the potential for "irreversible" damage to the denitrifying microbes in acidified soil. However in view of the high quality of the scientists who are members of this committee, we would be well-advised to consider their comments carefully. I look forward to the release of the complete report from this committee.

APPENDIX C: COMMENTS OF JACK G. CALVERT AND ALAN LAZRUS ON THE INFORMAL "REVIEW" OF THE NRC COMMITTEE REPORT ON ACID DEPOSITION SUBMITTED BY THE BERNARD MANOWITZ GROUP

Recently we received a copy of a "review" of our NRC deposition report which was prepared under the guidance of Bernard Manowitz (Brookhaven National Laboratory) by selected representatives of three national laboratories supported by the Department of Energy: Brookhaven, Battelle Pacific Northwest, and Argonne. These "reviews" were sent to the National Academy of Sciences, to me, to various members of the Interagency Task Force on Acid Rain and others; my friends in industry and government tell me that the report was widely circulated to the leadership and research arms of the power industry and related industrial operations throughout the United States and within the extensive communications pipeline of the administrative and legislative staffs of the Federal and State governments. The original report has subsequently been sanitized and presented to Representatives Fuqua, Scheuer, and Lloyd with additional comments from a group at Oak Ridge National Laboratory; this has been or will be introduced into the Congressional Record. We did not have access to the revised version of the "review" of the Manowitz group until a few days ago, so we will necessarily respond to the original "review" which has received wide circulation. The scientific questions remain unchanged in the latest version. I have submitted as Appendix D of the formal report for the Congressional Record, the original Manowitz group "review" which should be published as part of the record of this hearing. The comments given here represent those of only two of our NRC acid deposition committee, myself (Jack G. Calvert) and Alan Lazrus, since time did not allow the participation of the entire committee.

A Congressional hearing or the Congressional Record is not well suited to scientific debate. In the long run scientific issues cannot be influenced by

legal or legislative actions or by pressures from business, conservation groups, or other special interest groups, including that headed by Manowitz. The scientific world does not accept blindly the stated opinions of any scientist or group of scientists, including the NRC committee on acid deposition, without extensive testing and checking of the ideas proposed. Fortunately for science the unresolved issues will be decided after extensive research and suitable discussions in the refereed scientific literature. However, I feel that the Manowitz group and its informal "review" cannot be disregarded by me since their "review" has been presented to a wide and important audience as a "scientific" endeavor which disputes some key aspects of the NRC acid rain report. Although it is difficult to sort out the science from the rhetoric in some of these "reviews", we will focus on what we judge to be the important scientific questions which have been raised.

First, the use of the Hubbard Brook data by the NRC acid deposition committee and our conclusions derived from them have been questioned. A reviewer designated as "D" has attempted to treat these and other data not available to the general scientific community, using a multiple linear regression analysis of "adjusted" data. He concluded that there is no 1:1 correspondence between sulfur oxide emissions and sulfate deposition at Hubbard Brook. However his conclusions must be rejected since his methods of data treatment and his arguments are flawed scientifically. The detailed analysis of the errors in his treatment have been submitted as part of the record of this hearing along with the original Manowitz group "review". It suffices for present purposes to indicate that he has made two serious errors. He made an inappropriate choice of variables in the multiple linear regression analysis, and he "corrects" the observed data using a long-range model and assumptions about the source regions based upon "local" winds (measured 100 miles from Hubbard Brook on the coast of Maine) which are definitely incorrect.

Given the lack of reliable models to predict accurately the source regions for the receptor site of Hubbard Brook, it is not scientifically sound to attempt the treatment of emissions and sulfate concentration at remote sites given by reviewer "D".

The conclusion of the NRC committee, namely that a large decrease in sulfate deposition at Hubbard Brook corresponded in time with a large decrease in sulfur dioxide emissions in the Northeast, is as definitive a statement as was permitted by the data. Certainly the attempt to be more quantitative made by reviewer "D" is seriously flawed scientifically, and must be rejected. His treatment of the sulfate to nitrate ratios which appears in the revised version of the Manowitz group "review" suffers from many of the same problems, and the results are of uncertain quality.

Reviewer "E" questioned the near constancy of the $SO_4^{=}/NO_3^{-}$ ratios observed over wide areas of eastern North America in the annual averaged deposition data. His comments show such an extraordinary degree of rancorous disdain and distortion that one should not honor them with a response. However our analysis of his comments (placed in the written material for the Congressional Record) shows no scientific basis for his claim. Instead a serious distortion of the observations is evident.

Our conclusion that reliability for present long-range transport and transformation models is low in relating specific sources with receptor sites has been questioned by one of the Manowitz "review" group ("F"), yet apparently it is supported by others of this group. The objection is surprising since our view seems to be the consensus view of the NRC committee on acid deposition, the Interagency Task Force on Acid Rain (of which the Manowitz group is some undefined part), and the Acid Rain Peer Review Panel.

Models are used by all scientists today, and certainly definitive uses have been and will continue to be made of them. Our caution relates to their inappropriate use. If long-range modeling involving the attribution of specific sources to specific receptor sites is used today, and of course it is now used by some for this purpose, then the modellers should provide the user with realistic estimates of the magnitude of the uncertainty associated with the results so that these can be incorporated into the control strategy planning. Such a practice is not suggested to frustrate the control plan, but it is necessary to provide the sense of realism that is demanded in the formulation of sulfur oxide emission control strategy in view of the very high costs associated with various control strategy options.

Certainly the conclusions of the NRC committee on acid deposition are necessarily very limited in nature, and indeed they must and will be checked by extensive further experimentation. Our conclusions represent the best judgment of a group of highly qualified and diversified scientists, and we certainly would not alter or compromise these conclusions on the basis of the unusual "review" of the Manowitz group.

Jack G. Calvert
Alan Lazrus
Senior Scientists
National Center for Atmospheric Research
1850 Table Mesa Drive
Boulder, Colorado, 80307

BIOGRAPHICAL DATA FOR JACK GEORGE CALVERT

Born in Inglewood, California, U.S.A., May 9, 1923; married to Doris Arlene Breimon, November 8, 1946; children: Richard John (born February 23, 1955) and Mark Stephen (born December 27, 1957).

Present Residence: 260 Cactus Court, Boulder, CO 80302 (Tel.: 303-449-4160).

Business: Atmospheric Chemistry and Aeronomy Division, National Center for Atmospheric Research, 1850 Table Mesa Drive, P. O. Box 3000, Boulder, CO 80307 (Tel.: 303-494-5151, ext. 393; FTS 322-5393).

Higher Education: B.S. (Chemistry), University of California, Los Angeles, 1944; Ph.D., Physical Chemistry (Photochemistry) with Professor F. E. Blacet, University of California, Los Angeles, 1949; Postdoctoral Fellow, National Research Council (Canada) with Dr. E. W. R. Steacie in Photochemistry, 1949-1950.

Employment:

U.S. Navy, Apprentice Seaman-Ensign, 1944-1946 (Radar)
Teaching Assistant, Research Assistant, U.C.L.A., 1946-1949
Postdoctoral Fellow, National Research Council (Canada), 1949-1950
Assistant Professor of Chemistry, The Ohio State University, 1950-1956
Associate Professor of Chemistry, The Ohio State University, 1956-1960
Professor of Chemistry, The Ohio State University, 1960-1981
Chairman, Chemistry Department, The Ohio State University, 1964-1968
Kimberly Professor of Chemistry, The Ohio State University, 1974-1981
Professor Emeritus, The Ohio State University, 1981-present
Consultant in Kinetics, Photochemistry, Free Radical Reaction, Environmental Chemistry, several industries, institutes, agencies including: National Cash Register Company; Union Carbide Company; Environmental Resources, Inc.; Battelle Memorial Institute; General Motors Corp.; Stanford Research Institute; Environmental Research and Technology; Coordinating Research Council; Environmental Protection Agency (U.S.); Ministry of Environment, Ontario, Canada
Senior Scientist, National Center for Atmospheric Research, Atmospheric Chemistry and Aeronomy Division, 1982-present.

Honors:

Highest Honors in Chemistry, 1944, University of California, Los Angeles, with B.S.
State of California Scholarship, University of California, Los Angeles, 1944
H. M. Mosher Fellow in Chemistry, University of California, Los Angeles, 1948-1949
National Research Council (Canada), Fellow in Chemistry, 1948-1949
Honor Professor of the Year, College of Arts and Sciences, The Ohio State University, 1957
Alumni Distinguished Teaching Award, The Ohio State University, 1961
Frontiers in Chemistry Lecturer, Case-Western Reserve University, 1963
Visiting Lecturer in Physical Chemistry, University Western Ontario, London, 1973
American Chemical Society, Columbus Section, Treasurer, 1955; Secretary, 1956; Chairman-Elect, 1963; Chairman, 1964; Alternate Councilor, 1966;

Editorial Boards: Journal Photochemistry and Photobiology, 1966-1971; International Journal of Chemical Kinetics, 1967-1972; Reviews of Chemical Intermediates, 1978-
Kimberly Professor Chemistry, The Ohio State University, 1974-1981
Simon Guggenheim Memorial Fellowship, 1977-1978
American Chemical Society, Columbus Section Award, 1981
Distinguished Research Award, The Ohio State University, 1931
Fellow, American Association for the Advancement of Science, 1981
American Chemical Society Award for Creative Advances in Environmental Science and Technology, 1982

Member:

Phi Beta Kappa (elected Junior Year), Phi Lambda Upsilon, Sigma Xi, Pi Mu Epsilon, Alpha Chi Sigma, American Chemical Society, Ohio Academy of Science (Fellow), Air Pollution Control Association, American Institute Chemists (Fellow),
American Association for the Advancement of Science (Fellow)

Committees, National:

U.S. Public Health Service, Air Pollution Training Committee, 1964-1968; Chairman, 1967-1968
National Air Pollution Control Administration, National Air Pollution Manpower Development Advisory Committee, Chairman, 1968-1969; Adjunct Faculty Member, Research Triangle Park, North Carolina, 1969-1970
Conservation Foundation, National Advisory Committee, Chairman, 1969-1972
Gordon Research Conferences, Member of Board of Trustees, 1970-1972; Member-at-large of Council, 1968-1970; Chairman of Gordon Conference on Environmental Sciences: Air, 1981
Environmental Protection Agency, Air Pollution Research Grants Advisory Committee, 1970-1972
Chemistry and Physics Advisory Committee on Air Pollution, 1973-1976
Committee on the Evaluation of Atmospheric Nitrosamine Impact, 1976-1977
Committee on Evaluation of Innovative Research Programs, 1978-present
Environmental Chemistry and Physics Review Panel, Chairman, 1980-present
National Academy of Sciences
Committee on Health Effects of Air Pollutants, 1973-1974
Committee on Health Effects of Nitrogen Dioxide, 1973-1974
Committee on Atmospheric Aldehydes, 1978-1981, Chairman, 1979-1981
Committee on Transport and Transformation Chemistry in Acid Deposition, January 11, 1982-present, Chairman
National Air Conservation Commission of the American Lung Association, 1974-1975
U.S.A.-U.S.S.R. Joint Committee on Atmospheric Modelling and Aerosols, 1975-1978
Brookhaven National Laboratory, Department of Applied Science, Visiting Committee, 1976-1979
Executive Committee, Inter-American Photochemical Society, 1979-present
National Center for Atmospheric Research, Scientific Programs Evaluation Committee's Review Panel on Atmospheric Quality Division, Chairman, 1980

Jack G. Calvert and his research group have studied the photochemistry and reaction kinetics of many molecules which are important constituents of the polluted atmosphere: SO_2, CH_2O, HONO, etc. He has been concerned with the elucidation of the complex mechanisms which are operative in the chemistry of the polluted troposphere. His research publications (about 160 in number) relate to photochemistry, reaction kinetics, atmospheric reactions, and environmental chemistry. He coauthored with James N. Pitts, Jr., a book "Photochemistry" published by John Wiley in 1966 and translated into the Russian language and published by the Mir Publishing House in Moscow, Russia, 1969. At the National Center for Atmospheric Research, Dr. Calvert is Head of the Precipitation Chemistry, Reactive Gases and Aerosols Section in the Atmospheric Chemistry and Aeronomy Division. Research in the Division focuses on both laboratory and field studies of certain trace components present in the stratosphere and troposphere with the aim of understanding the mechanisms of transformation of sulfur dioxides and other important trace gases.

Mr. SCHEUER. We've had a number of members come in in the last couple of minutes. Could you just tell us very briefly and very succinctly what the conclusions of your group are.

Dr. CALVERT. Yes. Our conclusion, put in a nutshell, is that if you do cut back emissions in the emitting areas, like the Midwest or the Northeast or whatever, if you cut them back, we expect that you will find in deposition a reward for that cutback integrated over the area, you'll get a reward that will be almost proportionate to the cutback in emissions.

One conclusion: You won't be wasting your money if you do put controls in. One limitation that we state: That those controls may not be effective in representing a proportionate decrease in the very sensitive areas because our models today are not sufficiently adequate to make fine prediction about where the effects will be felt. We know they'll be felt somewhere in the Eastern United States, but maybe not in Hubbard Brook; maybe not in the Adirondacks.

Our expectation is it's not going to be a waste of money, though. It will certainly cut back in the Northeast United States somewhere.

Mr. SCHEUER. Thank you very much and I want to apologize again for beginning to exercise a little tighter rein on the hearings and it's because you've attracted such an excellent attendance of Congressmen.

All right, Dr. Ackermann, we're looking forward to your testimony very much. We've all read it; it's very thoughtful and stimulating. Can we ask you to sort of sum up in 8 or 10 minutes at the most so we can all have a chance to ask you some questions.

Dr. ACKERMANN. Yes. Thank you, Mr.——

Mr. SCHEUER. And pull that mike in, if you don't mind.

Dr. ACKERMANN. Am I on now? Yes.

Mr. SCHEUER. Pull it in a little closer so you don't have to strain.

Dr. ACKERMANN. All right.

STATEMENT OF DR. WILLIAM C. ACKERMANN, ADJUNCT PROFESSOR OF CIVIL ENGINEERING, UNIVERSITY OF ILLINOIS AT URBANA-CHAMPAIGN, AND VICE CHAIRMAN, ACID RAIN PEER REVIEW PANEL, OFFICE OF SCIENCE AND TECHNOLOGY POLICY, EXECUTIVE OFFICE OF THE PRESIDENT

Dr. ACKERMANN. Thank you, Mr. Chairman——

Mr. SCHEUER. Fine.

Dr. ACKERMANN [continuing]. Members of the subcommittees. Delighted to be here this morning to—as the Vice Chairman of the Acid Rain Peer Review Panel——

Mr. SCHEUER. Would you express our regrets and warm regards to Dr. Bill Nierenberg and tell him that we look forward to the day when we'll enjoy his wisdom here in person.

Dr. ACKERMANN. Well, thank you very much. I will certainly do that.

This Acid Rain Peer Review Panel, of which he's chairman and I'm vice chairman, was established, as you know, in the Office of Science and Technology Policy in the Office of the President, to give advice on the activities that were proceeding under the work-

ing groups established by the August 5, 1980, memorandam of intent on the transboundary air pollution between the United States and Canada.

We have organ—we have analyzed the results of those joint studies. With your permission, I will, indeed, summarize our findings briefly and ask that our full testimony be received.

Mr. SCHEUER. Let me say at this point that every—the full testimony of all three of the witnesses will be printed in the record at—just proceeding the point at which they started chatting informally with us. Yours will be printed in full in the record at this point.

Dr. ACKERMANN. Thank you.

[The prepared statement of Dr. Ackermann follows:]

Testimony of William C. Ackermann at a Hearing on
Acid Rain: Implications for Fossil Energy R & D
Held Jointly by the
Subcommittee on Energy Development and Applications
and the
Subcommittee on Natural Resources, Agricultural Research and Environment
of the
Committee on Science and Technology
of the
U.S. House of Representatives
September 13, 1983

Mr. Chairman, Members of the Subcommittees, Good morning.

My name is William C. Ackermann, an Adjunct Professor of Civil Engineering at the University of Illinois. I am pleased to appear this morning at your invitation as the Vice Chairman of the Acid Rain Peer Review Panel established by the Office of Science and Technology Policy in the Office of the President. Our Chairman, Dr. William A. Nierenberg, Director of the Scripps Institution of Oceanography, was unable to appear because of a previous commitment.

I have been asked to present the findings of the interim report of our Panel, and also comment on the National Research Council's recent report titled "Acid Deposition: Atmospheric Processes in Eastern North America" as well as on the "Annual Report, 1982," of the National Acid Precipitation Assessment Program.

My oral presentation will summarize a longer statement which is being submitted.

The Acid Rain Peer Review Panel was commissioned to review reports of the Working Groups established under the August 5, 1980 Memorandum of Intent (MOI) on Transboundary Air Pollution between the U.S. and Canada. We were to consider whether the Working Groups took into account currently available scientific and technical knowledge on the production, transport, transformation,

and deposition of pollutants; the effect of these pollutants on our surroundings, and the economics and engineering estimates of control technology performance. We were to provide an assessment of whether the Working Groups fulfilled their charters, utilized significant research and data, identified assumptions, provided support for conclusions and recommendations, and whether they addressed the uncertainties in available knowledge. The Panel was to provide an independent assessment of the uncertainties in available scientific and technical information, recommend further research and monitoring, and to provide a written report.

On June 27, 1983 the Panel released a brief report entitled "General Comments on Acid Rain" which is the basis for this testimony. A final report including specific comments on the MOI reports and on research is expected this fall.

The United States and Canada together are emitting annually more than 25,000,000 tons of sulfur dioxide (SO_2) and a comparable amount of nitrogen oxides (NO and NO_2, abbreviated as NO_x), and these oxides can be converted by atmospheric chemical processes into sulphuric (H_2SO_4) and nitric (HNO_3) acids. The emissions are large enough to increase appreciably the acidity of natural rainfall, and rain in most of eastern North America is considerably more acid than that expected from natural processes alone. The Clean Air Act of 1970 marked the formal recognition by the U.S. government of the importance of reducing the emissions of sulfur to the atmosphere, and new power plants constructed since that time do control such emissions to lower levels. Such controls were a prudent first step. We recommend that additional steps should be taken now which will result in meaningful reductions in the emissions of sulfur compounds into the atmosphere beginning with those steps which are most cost effective in reducing total deposition.

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The incomplete present scientific knowledge sometimes prevents the kinds of certainty which scientists would prefer, but there are many indicators which, taken collectively, lead us to our finding that the phenomena of acid deposition are real and constitute a problem for which solutions should be sought:

(1) The emissions of SO_2 and NO_x in eastern North America are at least ten times larger from human activities than from natural processes.

(2) A substantial fraction of such emissions are observed to return as sulfate ($SO_4^=$) and nitrate (NO_3^-) in rainfall; a probably comparable amount returns as "dry" deposition through surface interaction processes which are more difficult to monitor than the "wet" deposition in rain.

(3) In eastern North America the areas receiving the largest amounts of these acids are found within and downwind from the major source regions.

(4) The acidity of precipitation, and some streams and lakes in these major receptor regions are greater than the "natural" levels.

(5) Although some kinds of lakes have been acid throughout their known history, others located in principal receptor areas have become appreciably more acid during the past ten or twenty years.

(6) These changes in lake acidity have been accompanied by major changes in the biological activity within them, often including the disappearance of some species of fish.

(7) The largest of such aquatic effects have occurred in regions in which acidity is not "buffered" by the presence of alkaline minerals.

(8) Major areas of eastern North America have been identified whose geological composition is characterized by the absence of any important buffering capacity.

(9) Extensive evidence exists for increasing forest damage in eastern North America during the past few decades. Evidence of acid deposition as the primary cause for such harmful ecological effects on forests and other non-agricultural soils is, at present, much less compelling than that for aquatic damage.

The overall scientific understanding of the various aspects of acidic precipitation is quite incomplete at the present time, and will continue to have major uncertainties well into the future. Some of these gaps in our knowledge are permanent because the necessary measurements were not made ten, twenty, or fifty years ago before the potential future utility of such information was recognized. Other gaps exist because the needed scientific techniques have not yet been perfected or have not been adapted to the scale required for measurements covering much of the entire Western Hemisphere. Some of the important information will require at least ten or twenty years of additional data collection to take full cognizance of atmospheric variability and atmospheric cycles. Biological systems are extremely complex and variable. Response and recovery of many of these systems to external stress will require long-term (decades) detailed study for full evaluation. For these reasons, any current scientifically-derived recommendations must be based upon an imperfect, always increasing, body of pertinent data whose quality and completeness can be expected to improve for decades. Recommendations based upon imperfect data run the risk of being in error; recommendations for inaction pending collection of all of the desirable data entail even greater risk of damage.

The chemical processing of SO_2 and NO_x into acids in the atmosphere potentially involves a very large number of chemical reactions, and the relative importance of these various reactions changes drastically with time

and location, often in response to varying meteorological conditions. Sulfur and nitrogen can be removed from the atmosphere in various chemical forms, and by both dry processes at the surface and wet processes in rainfall. Measurements of $SO_4^=$ and NO_3^- in rainfall are now widespread, but do not have a long historical base. Measurements of dry deposition are so scattered (and experimentally doubtful) that quantitative assessment is essentially not possible even now.

The modeling of atmospheric emissions, transport and deposition has been confined almost entirely to the sulfur cycle, leaving nitrogen (and all else) to the future. The existing models do not agree with one another, and cannot be verified by comparison with observation because of the scarcity of good field data. They actually do not do very well in reproducing the observations on gaseous SO_2 that are available. Such models cannot be relied upon for (a) estimation of how much material emitted at A will be deposited at B; and (b) how much SO_2 will have been first converted to H_2SO_4.

There exists now no acceptable method for the determination of source/receptor relationships on a scale much smaller than "eastern North America." With a very large effort in laboratory atmospheric chemistry, in field measurements, and in atmospheric modeling, it might be possible within ten years (but certainly not five years) to produce a source/receptor model for eastern North America. We have great hope that methodology based on the use of natural tracers in fossil fuels may bypass some of these difficulties and perhaps reduce the time needed to elucidate this complex of problems. When a verified model exists in the future, there is a possibility that the source/receptor relationship will be sufficiently complex and variable that similar emission controls would still need to be assigned over rather large areas rather than locally.

Reduction below present SO_2 emission levels would reduce total sulfur deposition levels and as a consequence both reduce the probability for major changes in additional acid sensitive lakes or forests and allow the possibility for a return toward the original biological conditions existing in recently acidified areas.

The effects of acid deposition on biological systems in North America varies from certain to speculative. There is no question that some fresh water bodies have been altered in sensitive areas. The increase of acidity can reach levels which result in the release or "mobilization" of aluminum from solid minerals raising the possibility of toxic metal effects on biological species in both lakes and forest soils. There is strong evidence for damaging effects on limestone monuments, bridges and buildings, and other structures, but there is no good estimate of the economic magnitude of these effects.

The effect of air pollutants on agriculture may be important but the quantitative evidence is scanty. (An estimate for ozone damage to agriculture in the United States is five percent of the cash value. We anticipate that the overall effect of acid precipitation on crops could be comparably significant.)

There is a tendency in the scientific literature to speak of "long-term" and "short-term" effects, or of "irreversible" and reversible" changes. Damage to fresh water lakes, where it exists, may require a recovery time varying from a few years to tens of years when the stress is removed. This variation depends upon the availability in the environment of species for colonization, the extent to which trace element composition has been altered, and similar factors. The recovery time of a stressed sylvan environment is probably several decades or more in New England and Canadian latitudes. With both forests and lakes, the term "irreversible" might be used for a recovery time which stretches beyond a few decades.

We as a committee are especially concerned about possible deleterious effects of a sustained increase in the acidity of unmanaged soils. Its microorganism population is particularly sensitive to a change in acidity. But it is just this bottom part of the biological cycle that is responsible for the recycling of nitrogen and carbon in the food chain. The proper functioning of the denitrifying microbes is a fundamental requirement upon which the entire biosphere depends. The evidence that increased acidity is perturbing populations of microorganisms is scanty, but the prospect of such an occurrence is grave. It may take many years of accumulation of acidity, from wet or dry deposition, before measurable consequences would be observed. Such an effect is "long-term" or "irreversible." It may take at least that many years or longer for the soils to revert to their original condition. It is this possibility which provides us with the greatest concern.

"Acid rain" or acid precipitation belongs to a socially very important class of problems that have the superficial aspects of being amenable to a permanent solution achieved by a straightforward sum of existing technological and legislative fixes. This is very deceptive. Rather, this class of problems is usually not permanently solved in a closed fashion, but is treated more commonly to accommodate a steady increase in knowledge and understanding, taking various actions that appear most effective and economical at any given time.

It is in the nature of the acid deposition problem, that actions have to be taken despite incomplete knowledge. We have earlier given estimates of how long it may take to understand the "wet" chemistry, or the biological response. Reasonably accurate models incorporating relevant meteorology, chemistry, minerology and biology take even longer. If we take the conservative point of

view that we must wait until the scientific knowledge is definitive, the accumulated deposition and damaged environment may reach the point of "irreversibility".

We feel that the proper initial approach is to select particularly economically effective steps to begin to reduce our concerns in the light of gross transport and deposition features that have been identified, associated with seasonal and geographical variations. Purely as an example, it may be useful to consider having fuel of different sulfur content during different seasons since the efficiency for wet sulfuric acid deposition seems to be much less in winter. As other examples, first "least cost" steps might be gross reductions in sulfur emissions from non-ferrous smelters and intensifying coal washing.

I have also been asked to comment for the Panel on the 1983 report "Acid Deposition, Atmospheric Processes in Eastern North America." The report was prepared by the Committee on Atmospheric Transport and Chemical Transformation in Acid Precipitation of the National Research Council (NRC).

We consider the report excellent, and a fine complement to our own study. Although the NRC report is more limited in scope than the considerations of our Panel, the study is thorough, balanced, and sound.

In support of the congruence of the two studies, I will cite or paraphrase major and representative conclusions of the NRC report, all of which I believe our Panel is in agreement.

1. Ambient concentration of SO_2 and NO_x are much higher over much of eastern North America than can be accounted for by emissions from natural sources on a regional scale.

2. Concentration of sulfate in both the air and precipitation tend to reach their maximum values in summer in the northeastern United States.

3. With respect to meteorological processes the NRC report states that all sources in eastern North America must be considered as contributing in one degree or another to the phenomenon of acid deposition. Evidence exists for long-range transport of pollutants leading to acid deposition, but the relative contributions of specific source regions to specific receptor sites currently remain unknown.

4. With respect to atmospheric models the NRC report states that the actual limitations of predictive models for calculating the effects of sources on sulfate deposition are not well defined because of deficiencies in knowledge of atmospheric processes and the lack of coherent regional data on air quality and deposition. Current models have not provided results that enable us to have confidence in their ability to translate SO_2 emissions from specific sources or localized groupings of sources to influences on specific sensitive receptors.

5. In the near term, we believe that collection and analysis of field data are likely to lead to improved understanding more quickly than refinement of deterministic models.

6. Another issue of importance that has concerned the NRC Committee as well as our Panel is generally referred to as "linearity", which refers to whether changes in emissions result in proportional changes in deposition. On this subject the NRC report concludes that there is no evidence for a strong nonlinearity in the relationships between long-term average emissions and deposition.

7. Another issue concerns the influences of local and distant sources on receiving areas. We agree with the NRC report when it states that the relative importance for deposition at specific sites on long-range transport from distant sources as compared with more direct influences of local sources cannot be determined from currently available data or reliably estimated using currently available models.

8. The NRC report further states that because we cannot rely on current models or analyses of air-mass trajectories, we cannot objectively predict the consequences for deposition in ecologically sensitive areas of changing the spatial pattern of emissions in eastern North America, such as by reducing emissions in one area by a larger percentage than in other areas.

9. Our Panel and the NRC Committee agree upon the necessity for extensive laboratory, field, and modeling studies to establish the physical and chemical mechanisms governing acid deposition.

Your Committee has also asked for comments on the "Annual Report, 1982," prepared by the Interagency Task Force on Acid Precipitation.

The Interagency Task Force was established by the Acid Precipitation Act of 1980 (P.L. 96-294) to plan and manage the National Acid Precipitation Assessment Program. The 20 members of the Task Force are drawn from 12 Federal entities, the directors of Argonne, Brookhaven, Oak Ridge, and Pacific Northwest National Laboratories, and four Presidential appointees. The primary responsibilities of the Task Force are:

1. Plan and manage the National Acid Precipitation Assessment Program.
2. Provide annual reports on the national Program's progress.
3. Produce an annual interagency budget for the National Program.

4. Coordinate the National Program with the research and monitoring activities of the private sector groups, environmental organizations, States, and other nations.

5. Maintain an inventory of federally-funded acid precipitation research projects.

6. Disseminate research results and assessments of their implications.

The Task Force has nine working-level Task Groups for research categories and one for international activities.

The Annual Report for 1982 is excellent, and can certainly be recommended as a brief and well-organized treatment of the subject of acid precipitation. In addition to providing an organizational and management setting for the national assessment, it is organized according to the 10 disciplinary topics which concern its Task Groups. These are: Natural Sources, Man-made Sources, Atmospheric Processes, Deposition Monitoring, Aquatic Effects, Terrestrial Effects, Effects on Materials, Control Technologies, Assessments and Policy Analysis, and International Activities.

Within each of these topic chapters, discussion material includes a section on background which is essentially a state-of-the-art summary. This is followed by sections on research goals and activities, program accomplishments in FY 1982, and future activities.

The assessment program seems designed to yield successively better answers to the questions of emissions, transport, deposition, effects, and remedial measures with definitive bases for policy decisions by about 1987 or 1989. For the present the report is tentative or conservative in its judgments. For example, it states that "man-made atmospheric pollutants are probably the major contributors to acid deposition in northeastern North America." It

further states that "The approximate coincidences of the region of highest precipitation acidity with the areas of greatest sulfur dioxide and nitrogen oxides emissions provides circumstantial evidence to support this assertion." Stress is placed upon "the lack of documentation of the mechanisms linking cause and effect." In the area of aquatic effects where evidence is perhaps the strongest, it states that "The question of long-term trends in freshwater acidification also remains uncertain."

Thus, while the Acid Rain Peer Review Panel, for which I am attempting to speak, finds much in the report with which to agree, we conclude that there is substantial evidence of cause and effect relations, and that an incremental program of mitigation should be undertaken with the most economical measures.

With regard to proposed future research activities, we believe that the proposals of the 1982 Report include studies which are necessary and important. If there is any difference in the views of the Acid Rain Peer Review Panel it is one of degree. We recommend priority attention for investigations on the quantitative understanding of the effects on the total ecological system. Of special importance may be effects on microbial activity in soil and, through that, on the nitrogen cycle.

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Brief Biographical Sketch of

WILLIAM C. ACKERMANN

Dr. William C. Ackermann is a 1935 graduate of the University of Wisconsin in Civil Engineering.

Successive employment was in the Water Control Planning Department of the Tennessee Valley Authority from 1935-1954, Head of the Watershed Hydrology Section of the U.S. Department of Agriculture from 1954-1956, Chief of the Illinois State Water Survey from 1956-1979, and Adjunct Professor of Civil Engineering at the University of Illinois from 1979-present. From 1963-1964 he was in the Office of Science and Technology in the Office of the President.

Dr. Ackermann has been a member or chaired numerous committees of the National Research Council and scientific and engineering societies. He has been a consultant to agencies in the U.S. and abroad. He is a member of the National Academy of Engineering.

Dr. ACKERMANN. Mr. Chairman, the United States and Canada, together, are emitting annually more than 25 million tons of sulfur dioxide, SO_2, and a comparable amount of nitrogen oxides, NO and NO_2, which we abbreviate as NO_x, and these oxides can be converted by atmospheric chemical processes into sulfuric acid, H_2SO_4, and nitric acid, HNO_3.

The emissions are large enough to increase appreciably the acidity of natural rainfall. The rain in most of eastern North America is considerably more acid than that expected from natural processes alone.

The Clean Air Act of 1970 marked the formal recognition by the U.S. Government of the importance of reducing the emissions of sulfur to the atmosphere and new powerplants constructed since that time do control such emissions to lower levels. Such controls were a prudent step.

We recommend that additional steps should be taken now which will result in meaningful reductions in the emissions of sulfur compounds into the atmosphere, beginning with those steps which are most cost-effective in reducing total deposition.

The incomplete present scientific knowledge sometimes prevents the kinds of certainty most scientists would prefer, but there are many indicators which, taken collectively, lead us to our findings that the phenomenon of acid deposition is real and constitutes a problem for which solutions should be sought.

I will cite nine partial indicators to illustrate this point:

One, the emissions of SO_2 and NO_x in eastern North America are at least 10 times larger from human activities than from natural processes;

Two, a substantial fraction of such emissions are observed to return as sulphate and nitrate in rainfall. A probable comparable amount returns as dry deposition through surface interaction processes which are more difficult to monitor than the wet deposition in rainfall;

Three, in eastern North America, the areas receiving the largest amounts of these acids are found within and downwind from the major source regions;

Four, the acidity of precipitation and some streams and lakes in these major receptor regions are greater than the natural levels;

Five, although some kinds of lakes have been acid throughout their known history, others located in principal receptor areas have become appreciably more acid during the past 10 or 20 years;

Six, these changes in lake acidity have been accompanied by major changes in the biological activity within them, often including the disappearance of some species of fish;

Seven, the largest of such aquatic effects have occurred in regions in which acidity is not buffered by the presence of alkaline materials;

Eight, major areas of eastern North America have been identified whose geological composition is characterized by the absence of any important buffering capacity; and finally

Nine, extensive evidence exists for increasing forest damage in eastern North America during the past few decades.

Evidence of acid deposition as the primary cause for such harmful ecological effects on forests and other nonagricultural soils is at

present much less compelling than that for adequate damage—than for aquatic damage, excuse me.

The overall scientific understanding of the various aspects of acidic precipitation is quite incomplete at the present time and will continue to have major uncertainties well into the future.

Some of the important information will require at least 10 or 20 years of additional data collection to take full cognizance of atmospheric variability and atmospheric cycles. Biological systems are extremely complex and variable.

For these reasons, any current scientifically derived recommendations must be based upon an imperfect, always increasing body of pertinent data whose quality and completeness can be expected to improve for decades.

Recommendations based upon imperfect data run the risk of being in error. Recommendations for inaction pending collection of all of the desirable data entail even greater risk of damage.

Mr. Chairman, I sense very much that you would like to limit the initial testimony so that we might have more time for questions and discussion. I would like to respond to the chairman's request that I comment briefly on the other two reports and I will do that very briefly, the activities represented by the other two witnesses this morning.

The first of these I would comment on is the one chaired by Dr. Calvert of the National Research Council and we certainly feel that his report is excellent and a fine complement to our own study in the Office of Science and Technology Policy. Although the NRC report is more limited in scope—being primarily concerned with the atmospheric processes rather than the considerations of our panel—we think that his study is thorough and balanced and sound in their area.

Then I would like to comment briefly also on the report that we heard of first this morning, the annual report for 1982 of the Federal activity of the Interagency Task Force on Acid Precipitation. We think also that this is an excellent report, Mr. Chairman, and can certainly be recommended as a brief and well organized treatment of the subject of acid precipitation.

The assessment program outlined by Mr. Bernabo seems designed to yield successively better answers to the questions of emissions, transport, deposition, effects and remedial measures with definite bases for policy decisions by about 1987 or 1989. For the present, their report is tentative and conservative in its judgments.

If I may, I will stop at this point, far short of the complete comments, but would be happy to join in questions or discussion in any way that you pose.

Mr. SCHEUER. Well, thank you very much, Dr. Ackermann, and thanks to all three of you. I think you whetted our appetite and stimulated us and that's—the best proof of that is the extraordinary showing you got here with eight Members here—not just very many minutes ago—and seven Members sitting here now.

We're going under the 5-minute rule, I regret to say, as of now. Have you got that?

I'm going to ask each of you one question and I would ask you to restrict yourself to 2 minutes, so I'll go 1 minute over the 5 minutes.

If you can't finish—and I hope you will be able to finish—I'm going to ask unanimous consent that we hold the record open for 8 or 10 days for you to submit answers to other Members' questions and other questions we will submit in writing. OK.

But in order to give all Members a chance to ask questions, we'll try and stick very rigidly to the 5-minute rule.

I'm going to ask you, Dr. Ackermann, you stated in your testimony that some lakes have become appreciably more acid during the last 10 or 20 years. Now, that seems to conflict with Dr. Bernabo's statement that long-term trends in fresh water acidification remain uncertain.

How come you have such a greater degree of certitude than our friend, Chris Bernabo? And how do you detect with more certitude the long-term trends in lake acidity?

Dr. ACKERMANN. I think it's a matter of degree, Mr. Chairman. I don't think we're in conflict. The record of which lakes have become more acidic is open to both of us and to the entire scientific community.

I think that it's a matter of whether one recognizes that there are other lakes that are in the same circumstance and could reasonably be expected to become acidic and affect their biological communities living in them as well. And I think that perhaps our panel may have extended further than scientists often care to go in projecting the future trends, and that is that the things that have been happening and have—are in our total agreement, can be expected to continue and extend to other areas that are subject to the same atmospheric impact and have also the limited alkaline-buffering capacity that the lakes have shown—as in the lakes that have shown change.

I hope that's reasonable response to your question.

Mr. SCHEUER. Chris, do you want to address that question?

Dr. BERNABO. Well, I think Bill's——

Mr. SCHEUER. In other words, if long-term trends in acidification are sort of fuzzy, then you put us in a position of being a little silly in asking—in even seriously contemplating laying onto industry the need for making significant capital investments, and the fact is that we don't want to look silly. We don't want to lay on industry unnecessary capital expenditures. We don't want to lay on industry any capital expenditures that cannot be justified on a cold, hard-nosed cost-effectiveness analysis.

So when you say that long-term trends aren't really very evident and can't be analyzed or justified based on the empirical evidence, you sort of take the wind out of our sails.

Dr. BERNABO. Yes, I think——

Mr. SCHEUER. Where do we stand now in terms of the—of the—of exercising some cool judgment in saying we now know enough to move ahead? Now, Volker Mohnen for many years has been saying, we don't know enough and, therefore, we can't do anything.

That's the point of view that until you know everything, you can't do anything. Now, out at NCAR in Denver, for the first time, they're saying, we don't know everything, but we think we know enough to take that first significant move forward—it won't do everything, but that is justified on the basis of what we know, and we think that doing a significant amount now, not trying to achieve

perfection and Nirvana and not trying to achieve a risk-free, pollution-free atmosphere—we think there are things you can do that are sensible and are cost-effective and will not impose a burden on industry that isn't sensible by reasonable men.

Now, are we not at that point in your opinion?

Dr. BERNABO. Well, I think in response to the question which you've asked Bill and which I think he's answered, it is a matter of degree. You have to understand a little difference in role between the two gentlemen next to me and the committees that they had formed to provide assessments and our role to just continually improve the objective basis of information.

There's a line over which, when you ask the question, do we know enough——

Mr. SCHEUER. To do something, perhaps not everything.

Dr. BERNABO. To do something, right. You're dealing really as much with a value judgment as you are with any scientific judgment and of course, scientists like to——

Mr. SCHEUER. That goes without saying. That's what we're here for.

Dr. BERNABO. Correct.

Mr. SCHEUER. We're not scientists. We're making value judgments——

Dr. BERNABO. Correct.

Mr. SCHEUER [continuing]. By the dozens every day.

Dr. BERNABO. That's right. But our charge, again, is to provide the objective information to stop short of that value judgment. That's where, again, the policymakers have to come into play; there are a lot of factors beyond science which have to be put into this equation.

We're not saying there isn't evidence out there that there have been trends in this area; there is, but it is generally seen—if you look at it in the most conservative scientific sense—as being rather sketchy.

Now, we are putting into effect major efforts to clear that up. For instance, I mentioned we're going back and looking at the sedimentary history of a variety of lakes which will give us the information which we did not go out and collect in those days, but which we can go back and reconstruct how rapidly has this been occurring.

At the present time, I don't think anyone can tell you what is the rate at which this resource has been degraded and what are—how will we project that in the future? We're trying to rectify that so while we are at this moment saying the trends aren't clear, there's no question that some degradation has taken place. We agree with that. We state that explicitly. There's no question that research can help us retrospectively trying to determine what was the rate at which this resource is degradated.

Now, again, these other panels have the charge, and that's important, to look beyond just the science and give their best judgment of what's there, and that judgment implies more than science.

Mr. SCHEUER. All right. I'm going to have to stop you, Dr. Bernabo, because we've hit my 5 minutes.

Dr. BERNABO. OK.

Mr. SCHEUER. Congressman Ottinger.

We'll have some further questions for you.

Mr. OTTINGER. Dr. Calvert, have you talked to the Administrator of EPA about the implications of the National Research Council report?

Mr. CALVERT. Yes, two times.

Mr. OTTINGER. What appears to be the process that the Administrator is following in consulting with the scientific communities on this issue?

Dr. CALVERT. It seems to me that he's talked to conservation groups; he's talked to business; he's talked to scientists; he's trying his best to try to get a feel for the problem and what the possible solutions are. My contribution has been minimal, certainly, but I've tried to express to him in words that were perhaps better phrased something of the—our report, and what it's findings were.

Mr. OTTINGER. I take it that in matching up—the problem of matching up sources of pollution and deposition is one that is so complex that we may—we certainly won't be able to resolve that problem in a short period of time and may not be able to solve it at all and that your basic position is that what goes up surely must come down—greatly oversimplifying it—and that the problem is a real one and we ought to get started on it.

Dr. CALVERT. You've summarized very well what I said, rather less succinctly.

Mr. OTTINGER. And, Dr. Ackermann, I take it that your position is that we don't know enough to make it worthwhile to take action at the present time, that you feel that a good deal of research—additional research—needs to be known before——

Dr. ACKERMANN. No, Mr. Ottinger, quite the contrary. Our acid rain peer review panel believes that the problem is very real, is very important, and that some further steps should, indeed, be taken now. We—we recommend starting with the most economical steps and we think that these steps should be incremental. There are uncertainties and in view of those uncertainties, we think that some first steps should be made and then we should be prepared to make midcourse corrections as we go along.

But our panel is recommending that we do, indeed, begin to further reduce emissions.

Mr. OTTINGER. Has any work been done yet on the costs and benefits of taking the kinds of actions which Congress is contemplating, what the costs of taking 10 million tons out versus taking 12 million tons reduction would be? Anybody on the——

Dr. ACKERMANN. I think that, as it often happens in benefit cost analysis, it's much easier to estimate the costs—in this case, the cost to the persons who are the emitters—than it is to evaluate the benefits. We'll always have great uncertainty with the benefits.

And I think that it would be our view that we do not have economic evaluations of the benefits. Whether you're talking about the ecology of lake systems and streams; you're talking about human and terrestial animal health; you're talking about transportation; you're talking about effect on materials like concrete and bronze and so forth; we do not have economic evaluations.

But we are reluctant to see the process go forward because we think that the—that the implications at the present rates of emissions are very serious.

Mr. OTTINGER. Dr. Calvert?

Dr. CALVERT. Yeah, could I respond to that very briefly? When you have friends, indeed, they try to keep you advised and I have a couple of friends that have always sent to me things that I didn't ask for but nevertheless are useful.

David Streets, at Argonne National Laboratories, has just done a report that you undoubtedly will receive a copy of, that is a rather effective analysis, of the Argonne National Labs has one of the better groups that do computer simulations of various models and cost-effective analysis.

The group as well that you should investigate, I think, is Dr. Fay and his energy laboratory at MIT. They have done some cost-analysis kinds of things. Of course, our Federal Government—EPA has done a great deal on this, as I understand it, and I think you have no end of advice.

But I would add to my statement that as you get this advice, keep asking, all right, what's the uncertainty of these numbers, and they start to overlap on certain control policies when you include those uncertainties.

Mr. OTTINGER. Well, there seems to be a good deal of concern about how far we should go, how fast. We understand that the Administrator of EPA may propose a solution that calls for an immediate effort to reduce the depositions by 4 million tons, perhaps targeted to a limited number of States to achieve a reduction of acid deposition in the Adirondacks. Doesn't that solution fly in the face of the NRC's conclusions that source receptor relationships are not well enough known to predict such emissions?

Dr. CALVERT. I don't think there's a conflict there. I do think that there is an attempt on our part to suggest that if we do cut back, we'll find what the result will be. We'll find that it does integrate over the area to be a cutback that was commensurate with the emissions cutback.

But we will find out if we do cut back in specific areas is what effect that does have, then, through Chris' studies, to find out what effect that did have on the Adirondacks. If it isn't like we suggested; we haven't committed at least the world to an economic discomfort.

Mr. OTTINGER. Are there—this is one last—maybe you can do it for the record—are there costs, you say, the benefit figures are very hard to come by—are there cost figures that would show the difference between an effort to reduce 4 million tons, 10 million tons, and 12 million tons would seem to be the principal figures——

Dr. CALVERT. This kind I have here, I think——

Mr. OTTINGER [continuing]. That are being bandied about?

Dr. CALVERT. Is common knowledge and there are many other groups. I am sure I'm being unfair to just pick out two, but people like to do this. When they do it, I think you have to use a little bit of jaundiced reserve and say, "Well, how accurately is this done?" You get an economic modeler, he uses some kind of physical model

which may be unrealistic in doing it. He doesn't know how accurate it is.

But there are plenty of studies and you will get a potfull of them as soon as you start asking.

Mr. OTTINGER. Well, we're asking. If the witnesses could provide for the record whatever information is available on that, I think it would be helpful.

Thank you, Mr. Chairman.

Mr. SCHEUER. Congressman Boehlert of New York.

Mr. BOEHLERT. I'd like to ask all the panel, if I may, if you would characterize 1983 as a breakthrough year? Are we finally over the big hurdle? I mean, is it universally accepted right now that acid rain isn't created in Heaven by divine providence; that it's manufactured here on Earth by man?

Would you say that this is the year when we finally acknowledge that?

Dr. BERNABO. I'd say that is, in fact, the consensus of the scientific community, but also, as you've heard——

Mr. SCHEUER. Pull the mike in, Chris.

Dr. BERNABO [continuing]. There is considerable consensus. It's one thing to diagnose an ill; it's another thing to prescribe a cure and the prescription of the cure, I think, is where much of the uncertainty lies.

Mr. BOEHLERT. Yes, but we finally acknowledged the source of the problem.

The other thing I would like to ask: Quite frequently, those of us who are sort of diehards on the issue, and we're mostly located in the Northeast, you know, pushing for progress and progress retroactive if possible—but quite often the detractors say to us, "Go slow, United States, because it really doesn't serve your interest all that much if you proceed pellmell and the Canadians don't."

And the argument has been given that the Canadians aren't as enthusiastic about this problem as we are. Would you, particularly Dr. Bernabo, comment on that?

Dr. BERNABO. Are you saying that the Canadians are not as enthusiastic about addressing the problem?

Mr. BOEHLERT. Yes, are they backing up their words with deeds? What's their commitment in terms of resources from the National Treasury, that type of thing?

Dr. BERNABO. I'm really not in a good position to speak for the Canadians, but all contacts I have is both in terms of the research and in terms of the policy, they are very serious about this issue.

Mr. BOEHLERT. But you know what we're spending and, I mean, you've given us the chart; are they spending comparable amounts?

Dr. BERNABO. Yes. In fact, it's not widely known, but as I said—I mentioned during my testimony, the Canadians are spending, between their provincial governments and their Federal Government, about $20 million a year in research, which per capita is 10 times——

Mr. BOEHLERT. A lot more than we are.

Dr. BERNABO. That's correct.

Mr. BOEHLERT. So 10 times as much per capita.

Dr. BERNABO. Per capita, yes.

Mr. BOEHLERT. And the final observation I'd like to make to you, you indicated that 10 percent of the Adirondack lakes were in critical condition. I would put it beyond that. I'm told that 212 Adirondack lakes are actually termed "dead," and that 256 probably require hospice care, so that is very serious.

I would hope you would acknowledge that.

Dr. BERNABO. Right. Well, I say again, there are 2,000-some-odd lakes and ours—it's a lot more useful—instead of talking about the number of lakes to talk about the surface area because of the small lakes—many small lakes are more sensitive than larger ones.

The term I used—I gave the 4-percent term "critical." That term "critical" is by definition of the State of New York. It's not by definition, as was the data.

Mr. BOEHLERT. And finally, the first critical assessment; you say it's due shortly. Do you—do we have a timetable on that?

Dr. BERNABO. Well, I think it's in its final stages of public review. It should be distributed within the next few months. And again, that's something which 54 scientists from around the world have been involved in. It should be a quite useful benchmark. It's a rather lengthy document, as you can imagine, but it is one which will really sort of provide a benchmark of what the status of the science is so as we go along, we can see what improvements we have or have not made.

Mr. BOEHLERT. Thank you. No further questions, Mr. Chairman.

Mr. SCHEUER. Thank you. Congressman Buddy MacKay of Florida.

Mr. MACKAY. Thank you, Mr. Chairman.

I have a question which may be even more simplistic than any that's been asked thus far. What is the downside risk in America committing itself to a program now of a reduction of 50 to 70 percent of emissions, both of sulfur and nitrogen? What is the chance—or what harm would we do if we have overcommitted on the basis of insufficient data?

Dr. ACKERMANN. Well, if I might respond to that, Mr. MacKay, we are emitting great amounts—you know, I mention in my testimony 25 million tons of sulfur products and perhaps equal amounts of nitrogen products. We're so far above the natural levels that I think there's very little chance that we will reduce too much.

But on the other hand, there are uncertainties about the economics of making such reductions, and if our panel's recommendations were to be followed, we would suggest making only those initial, most economical steps so that we don't run the risk of overshooting the mark, as you say in your question.

We think that that could indeed be avoided by making small incremental steps in checking on the effects of those steps.

Mr. MACKAY. I—I'm concerned that we think only in short-term measures and we define "long-term" as 20 years. Typically, the end of our career is long term.

It's amazing to me that we could be saying in one breath that the amount that we are emitting is 10 times more than nature and in another, we're saying, "But we don't want to react too strongly because we might spend more money than we need in the long term."

You're saying that the evidence that we have now doesn't cause you concern; that we don't even take into account long term. What about a century?

Mr. ACKERMANN. I think we do indeed need to have a long-term point of view, and as a matter of fact, our panel is—has its greatest concerns in some areas like the effect on micro-organisms and bacteria in the soil who are working for us to convert these minerals if we don't acidify the soil to the point that they can't live and work.

We are interested in the long-term effects—greatest, however, we're talking about an emerging technology where we've only had measurements for about the last 3 years. We started this atmospheric program about 1979—the measurement of it—and in that, only for the sulfur products that are—that are falling down in the wet rain and snow.

And so, there's a major uncertainty in the fact that perhaps three-quarters of the material to which we're at risk is not even being measured and we do need to be somewhat conservative.

Mr. MACKAY. Well, I—I thought I read that in your report and I'm simply trying to define what would be a conservative response and it would appear to me that you're taking a very conservative view toward what would be a conservative response, almost like saying the main risk is we might spend more money than we needed to.

Dr. ACKERMANN. No, I don't think so. I don't think we would spend more money than we need to. We—it's a long-term problem, but we have a short-term technology and so we should take some steps, some cautious steps, be prepared to change the rate and direction and location of future steps as we see the effect of what we do, but we think the time—and if 1983 is indeed a year of breakthrough—it may well be that we at least—we have finally said that the problem is real—is real and present and we're going to start to do something about it.

Mr. MACKAY. I wouldn't want to come across as unduly negative. I think your report was very timely and I congratulate the three of you for having gotten us this far.

Thank you, Mr. Chairman.

Mr. SCHEUER. Congresswoman Schneider of Long Island.

Mrs. SCHNEIDER. Mr. Chairman——

Mr. SCHEUER. Of Rhode Island.

Mrs. SCHNEIDER [continuing]. At the outset, I would——

Mr. SCHEUER. Go ahead.

Mrs. SCHNEIDER. Thank you. I'd like to ask the opportunity to include in the record an article that was printed in the Providence Journal newspaper in the State of Rhode Island outlining the involvement of the State in their own survey of the lakes and ponds and I wonder if I might have permission to do that please?

Mr. SCHEUER. That's a single article?

Mrs. SCHNEIDER. Yes. Very brief.

Mr. SCHEUER. Without objection, so ordered.

[The newspaper article follows:]

[From the Providence (R.I.) Journal, July 10, 1983]

Fear of Losing Ponds Puts R.I. on Track of Acid Rain

(By Lester Boyd)

The water in the pond is clear with a faint, blue-green tint, much like the sterile water in a swimming pool. The only vegetation is the moss on the bottom.

The pond is dead.

The fish, the amphibians, the acquatic insects, the plankton, all the elements that make a pond a self-sustaining ecosystem are gone. They were killed by acid rain.

You can find ponds like this in New York, Pennsylvania, eastern Canada and Europe.

You cannot find ponds like this in Rhode Island, but the danger is there.

And the fish, says Andy Demaine, "may be just the first warning."

Demaine's business is fish.

He's a senior fish biologist for Rhode Island's Division of Fish and Wildlife, and his immediate concern is the effect of acid rain on the state's streams and ponds.

"We can measure the acidity in the water, and we know what levels of acidity the fish can tolerate. We can treat a pond with lime to neutralize the acid. But the fish may be just the most visible aspect of the problem. There's evidence it can also kill trees, and we have no idea what its ultimate effect on humans will be," he said.

An interim report on a Division of Fish and Wildlife study warns: "If acid deposition continues at the present rate there is a strong possibility that by the turn of the century waters in the rocky, thin-soiled region along Rhode Island's western border will, at best, be reduced to populations of redfin pickerel and yellow perch or, at worst, be devoid of aquatic life."

Andy Demaine has growing evidence that some of the state's waters are reaching the point where aquatic life could be wiped out.

"When we started," he said, "we grouped the ponds and streams together by watershed, but now I think that was the wrong way of doing it. It should be done by geologic area because the geology of a region seems to determine whether a pond or stream has a natural buffering capacity."

Soils and rocks with a high alkaline content are able to neutralize the acidity in the water. And ponds and streams fed largely from underground sources rather than from surface runoff tend to have a greater buffering capacity.

The acidity of water is measured on what is called a pH scale. On that scale seven is neutral, neither acid nor alkaline. Lower readings indicate acidity, and the acidity is logarthmic. That is a reading of six to 10 times more acidic than seven while a reading of five is 100 hundred times more acidic than seven.

Anything below a reading of five is dangerous.

Some fish are more vulnerable than others. Smallmouth bass cannot breed if the pH is between 5.5 and 6. Golden shiners will not reproduce at 4.8 to 5.2. Of the three species of trout found in Rhode Island, rainbows have the lowest tolerance for acid water. They can't breed if the level drops below 5.5. Brown trout tolerance is 5 and brook trout fail to reproduce when the acidity falls below 5.

If the fish cannot reproduce, no new generations come along and eventually the fish population dies out.

If the pH reading falls another half point below those listed above, chemical changes in the bodies of the adult will kill them.

There have been cases where the acid level in a body of water was such that rainbow trout could not be stocked. Last year there was a die-off of rainbow trout in the Ponaganset River in Glocester.

Normal rainfall has a pH of about 5.6. However, month-by-month monitoring of rainfall in Rhode Island shows nothing higher than 4.7 over the last two years, and some rain was as low as 3.7 on the pH scale.

In their monitoring of Rhode Island waters, biologists are looking beyond the pH reading, they're looking at the capacity of the water to neutralize the acid—known as buffering.

"That's the key to it," Demaine said. "When we're done we'll know which waters we have to be concerned with and which can take care of themselves.

The capacity of a body of water to neutralize acid is measured by the amount of alkalinity in a liter of water. A pond or stream with a reading of 2.5 milligrams per liter or above is able to neutralize the acid in rain. Any reading below that indicates the water is sensitive, and has little capability to neutralize acid.

Many state waters do not have the ability to neutralize acid. In some, the results are already showing.

For instance when Cedar Swamp Brook in Charlestown was sampled in the 1960s it had a population of native brook trout, as well as other species, but when it was sampled in 1982 only one brown bullhead, two eels and 16 small redfin pickerel were found. Brook trout had disappeared completely. The brook showed a pH reading of only 4.22 and no buffering capcity when tested last year.

The standard treatment for high acid water has been to add lime, and two bodies of water have required treatment. Wallum Lake in the northwestern corner of the state, prime trout water, and the pike rearing pond at Burlingame both have been limed.

The Burlingame pond is used by the Division of Fish and Wildlife to rear pike from fingerling to stocking size—about 12 inches. Demaine said when alewives were introduced to the pond to provide forage for the pike, they all died because of the acidity.

He said the division intended to introduce alewives into Beach Pond as forage for game fish, but tests showed the water is sensitive to acidity, so the plan was abandoned.

Treating a body of water with lime is a temporary solution only, and it can be expensive. According to John Cronan, director of the Division of Fish and Wildlife, it costs from $150 to $200 an acre to treat a pond.

The 37 locations sampled in the Pawcatuck River watershed show several ponds and streams with a pH of 5 or below and little or no buffering capacity, but the Queens River, Beaver River, Wood River, Flat River and Falls River, all in the same watershed, have pH readings between 6.25 and 7.

Of the ponds tested, Chapman's Pond in Westerly is the least sensitive to acidification. The fact that a landfill operated adjacent to the pond until the middle of 1970s may contribute to that.

Ponds and streams on the easterly side of Narragansett Bay were generally more resistant to acidity than those on the Westerly side of the Bay. But the project still has a year to run, and it won't be until all the figures have been analyzed that biologists will have a clear idea of what is happening to Rhode Island waters.

When the full picture finally emerges, biologists will know which ponds and streams will support populations of trout. This knowledge will serve as a guide for the trout stocking program. Waters that are sensitive to acidity will be taken off the list of stocked waters, or they will be treated with lime.

Demaine also came across something else that may be significant, but he admits he doesn't know why it happens.

Those ponds and streams where there is human involvement show better buffering capacity.

"If there are camps or cottages around a lake, or if there are farms, or any kind of human involvement, we get better readings."

"This puts us between a rock and a hard place." he said. "Human involvement pollutes, but it may be helping in this respect."

Also, what high acidity in drinking water will do to humans is a concern of health officials.

Dr. Jason Weisfeld of the state Department of Health said high acidity in the water is a potential health hazard. He said there is some evidence the acid causes heavy metals to leach into the water, and this could cause neurological disorders such as those associated with lead or mercury poisoning. And the heavy metals can reach humans through fish they eat from high acid water. "We're keeping our eye on the situation, and we're watching the literature for anything that might indicate we should be more concerned," he said.

Until action is taken to reduce the amount of acid in rainfall, waters in the Northeast will continue to be threatened.

The state, Demaine says, can only identify problems and deal with them on a patchwork basis.

Mrs. SCHNEIDER. Which leads me to my first question, that in the study that was done in 1982, the September study, Mr.—Dr. Bernabo, I wonder if you could tell us to what degree the non-Federal sector had been active in the research and the monitoring of acid precipitation?

Dr. BERNABO. Yes. I think—I'd like to just correct one misconception which might have come from my colleague, Dr. Calvert's testimony. The national program does not just include people from National Laboratories and Federal establishment. The fact there is

substantial involvement, although universities probably won't—there is substantial involvement of university scientists, State and local governments, and others.

In the monitoring activities, in fact, the primary performers of that work is the Illinois State Water Survey. They are the ones that serve as central coordinator of samples all over the country and—during the analysis.

So we have involvement of a number of groups. We do not have, to my knowledge, efforts such as you're describing in Rhode Island of volunteer involvement and so forth, although in some cases, there may be merit to that.

The problem is in getting the standardization when we found through long and hard mistakes that there can be very often uncertainties introduced into the data collection if it's not done in a very rigorous way, which is sometimes hard to do with volunteers.

On the other hand, I think it is very laudible that citizens become involved in that manner. There is substantial involvement outside the Federal agencies in the program.

Mrs. SCHNEIDER. OK. The other thing that I was interested in addressing is that in 1980, the budget for dealing with acid rain was $11 million. In 1984, it's $28 million, and I wondered if you could tell us a little bit about how that money is distributed among the six participating agencies.

Dr. BERNABO. Yes. I had shown a pie chart during my presentation——

Mrs. SCHNEIDER. Ah——

Dr. BERNABO [continuing]. I'll be happy to provide that in tables. Basically, though, about one-half of the total funds are flows through the Environmental Protection Agency, and the remaining eighths proportions flow through the Department of Agriculture, Interior, Energy, and NOAA, each with roughly equal amounts.

Mrs. SCHNEIDER. I thank you. Could you—would you feel that it would be safe to say that given the dramatic increase from $11 to $28 million, the quantitative amount of research that has been accomplished is significant?

Dr. BERNABO. Yes, I think the rate of the outputs since 1982 was the first year we had a concentrated program and expanded budget, the amount of outputs—it's like sowing seed, it's going to increase quite substantially as we go on. But it's also, I think, important to caution that no matter what the spending levels, a certain rate of good science can be done.

So I think the outputs——

Mr. SCHEUER. Excuse me, we didn't hear that. Would you pull the mike a little closer?

Dr. BERNABO. Yes, sorry, sir.

I'm saying that the outputs of the program, now that we've started in 1982 and 1983—we're now finishing up our second year—are going to be coming at a much more rapid pace as the more dollars and so forth have come into the program.

Also, the program has become much more focused than it was in 1981 when it was a bunch of piecemeal approaches, rather than trying to integrate the information. But I did want to caution that despite the increases, I'm not promising that we're going to answer all the questions. I think just because of the nature of the science,

that is not the case, but we'll be in a lot better shape as we move on than we have been.

Mrs. SCHNEIDER. I think that that's one of the concerns of many Members because we're looking toward the fiscal year 1984 budget and what kind of request do you anticipate making to us on that?

Dr. BERNABO. Well, the figure that you quoted, $28 million, is the 1984 request. This current fiscal year 1983, we're at $22 million.

Mrs. SCHNEIDER. OK.

Dr. BERNABO. We have requested about a 24-percent increase in the budget.

Mrs. SCHNEIDER. And are we to anticipate continuous requests for increases? I think we are concerned that given the amount of data and the lack of enthusiasm on the part of the administration for moving toward a definitive plan of action is rather discouraging, to say the least.

Dr. BERNABO. Well, I think you'll see, and again, it's premature to discuss, but I think as the administration—the effort which Bill Ruckelshaus has underway now comes forward, you will see that research is an integral part, I think, of any proposed approach.

As far as whether we'll be continually requesting more money, again you have to understand, we started from a very low base in the sense that there was very little research going on. I don't think we'll be continually asking for more, although this growth—we've tried to accommodate growth as best that can be done in an efficient management way, given the wide range of issues that raise.

Also, as we've probed into the issue, we've found it more complex than we believed it would be.

Mrs. SCHNEIDER. Well, when you mention the wide range of issues that are involved, one that I would like to add is that I serve on the Select Committee on Aging and we had recently held a hearing—sorry, I'm losing my voice here—which had made the linkage between Alzheimer's disease and the high aluminum content that is found in the blood of Alzheimer's victims, believed to be connected with acid rain.

And I was wondering if the Environmental Protection Agency is involved in some independent research of their own in this area, or——

Dr. BERNABO. Well, we are——

Mrs. SCHNEIDER [continuing]. If you are familiar with this.

Dr. BERNABO. We are looking at the——

Mrs. SCHNEIDER [continuing]. Discovery.

Dr. BERNABO [continuing]. Release of aluminum and other metals in water supplies. However, it is my understanding—I'm not an expert in this area, but that the relationship between the aluminum levels in the bloodstream and the input to the body is not well established in the first hand.

The second hand, things like using aluminum cookware and so forth may contribute far more than the current levels of drinking water. So I don't think there is any clear relation yet established. However, there's certainly reason for concern.

Mrs. SCHNEIDER. That's interesting that you should mention that because the panel of medical doctors had clearly indicated that it would be possible to eat from aluminum cookware for many, many, many years and not at all come close to the amount of levels of

aluminum being found in some water supplies. So it's just the complete opposite of what you're saying.

Dr. BERNABO. I haven't seen the outcomes of your hearing, but obviously I should take a look at those.

Mrs. SCHNEIDER. And as a member of this committee, I would like—and one who is also concerned with public health and right now my own voice—I would like to encourage your agency and your department to pursue the public health risks that are involved with acid rain because we've talked about the detrimental impact on agriculture and forest and the Brooklyn Bridge and everything else, but I think that we certainly have to consider human health as one of the, if not the highest, priorities.

Dr. BERNABO. Yes. I think that's a good point and again, I'd like to mention that we have underway right now in all the northeastern United States, a major survey of drinking water supplies and the levels of aluminum and other elements.

Mrs. SCHNEIDER. Thank you, Mr. Chairman.

Mr. SCHEUER. Thank you.

On the question of acid rain research, at least on this one question, one has to credit the administration. They have at least doubled the acid rain research since they have come in.

Congressman Mike Andrews——

Mr. ANDREWS. Thank you, Mr. Chairman——

Mr. SCHEUER [continuing]. From Texas.

Mr. ANDREWS. Dr. Ackermann, you, in your written statement, go into some detail about lack of adequate measurement devices to determine the extent of acid rain deposits on forests in the Northeast, and I wonder if you would expand on that a little bit and maybe enumerate what steps you think need to be taken to have better measurement criteria so we can have better data to make long-range recommendations?

Dr. ACKERMANN. Well, it's indeed a complicated problem. Back in my earlier time, I was responsible for evaluating the effects of a—copper smelters down in southeast Tennessee in the copper basin, and in that case, it was the direct sulfur dioxide emissions when those operations down there that killed the vegetation by the contact from the sulfur dioxide in the air to the leaves.

The process isn't always that simple. As these—these sulfur precursors move through the system, they may convert to sulphates, but they aren't necessarily converted into the liquid form in rain or snow. They may stay as a gas in the air, and they may also be an aerosol, but that's thought to be less—a very small particle.

Say a gaseous effect. It's very difficult; it's not like putting a milkcan out in the field and seeing what drops into it because we could do that with some refinement with the liquid part, but the gaseous part is subject to the winds and currents that move. It's also very much a factor of the kind of surface. If we have a smooth, snow surface, this is one thing. If we have a tree with some kind of a cone or a needle and leaf structure, it will collect differently than a flat surface, and so how the people who are most intimately involved are going to solve this question, I'm not sure.

But somehow or other, the—the different kinds of surfaces have to be simulated in the measurement technique and I think that we will not have a complete picture of—of the pollutant that's attack-

ing us, seriously, and particularly as it applies to vegetation because it's the surface that is affected, and of course, there are downstream effects, too, as these dry gases collect on the vegetation and may drop off or wash off, get into the soil and secondary effects like the aluminum release becomes effective.

Mr. ANDREWS. Doctor, how imprecise is the data?

Dr. ACKERMANN. I—we have been collecting—you mean the dry deposition. We have a standard collector. It was developed at the Illinois State Water Survey when I was chief there—for a wet collector on one side and a dry collector on the other, and the lid moves as it rains or does not rain.

And I was initially under the impression that this was a reasonable measurement technique, that the people who were working most closely in this area now do not trust it at all. They do not use the dry collectors, as I understand it. And the methods that are used for estimating the dry deposition are based on theoretical formulas of atmospheric exchange between the atmostphere and the surface of the ground based on a lot of complex factors like wind velocity and moisture and elevation from the surface and so forth.

It's a—it's a theoretical computational procedure and I think that it has very little credibility.

Mr. ANDREWS. Thank you.

Mr. SCHEUER. Congressman Tom Lewis from Florida.

Mr. LEWIS. Thank you, Mr. Chairman.

Dr. Calvert, you make—you stated that the success of control strategies built upon current unsophisticated models of long-range transported acid deposition is unpredictable at best. To what extent, then, could you justify substantial investment in emissions equipment to abate this problem?

Dr. CALVERT. I think that my suggestion—personal suggestion—this isn't speaking for my committee, would be that, indeed, I would take the best available advice on models at the moment and see what they suggested.

I would take that advice and in addition ask for an assessment of what the uncertainties are in that prediction. I'd use that to try to do something fairly quickly. I would say that that judgment should be tested because that judgment could be in error as models now are not adequately tested, nor do we have the proper basis to test them. We haven't a backlog of measurements in the field for a number of years under a variety of conditions that would enable us to build those models with the precision—the empirical models—with the precision that would be accepted by the scientific community or by the engineer or by the taxpayer who has to spend a bundle on doing this.

So I would say, even though we have uncertainties, 20 years from now, you ask us these same questions, hopefully I could be available to answer them at that time. If you were to do that 20 years from now, we'd say, "Well, we've found out a lot in those 20 years, sir," but we don't know yet what is the responsible action that one should take because we don't know yet what the source/receptor relationships are.

My friends—if I may take just another second—my friends, for example, who are mathemeticians say, "What are you guys doing; you can't ever have a model that predicts source/receptor relation-

ships." Nature is a variable that cannot be predicted by mathematics.

On the average, you can predict, but for a given event, there is no way that one can predict what would be the deposition at a given site, from a given source, with an accuracy of 100 percent. It's just impossible.

So facing that end, that you can't do it right anyway, at the moment, and maybe in 20 years, why don't we do something now and find out what is right?

Mr. LEWIS. But, doctor, what are we going to do? You know, we're talking about the taxpayer and the consumer footing the bill in the majority of cases.

Dr. CALVERT. Yes.

Mr. LEWIS. We have to have a little bit more specificity. You just can't expect and say well, we'll go out and spend a good $100 million or $200 million for this and expect the taxpayers to pay for it and then come back and say, "Hey, we guessed wrong." We've got to have a better——

Dr. CALVERT. That's right.

Mr. LEWIS. Situation than that.

Dr. CALVERT. I think that the plan that—I'm speaking personally because my committee, obviously, weren't charged to face this issue. My personal feeling is that indeed if one does something, then that something should be checked on; it should be done in a fashion which I think all of us have intimated would be a realistic one; that we do indeed make some significant changes and that we find the result of those significant changes; we do so on a scale that's not going to bankrupt the taxpayer.

Mr. LEWIS. I still think, well——

Dr. CALVERT. All right, let me go one——

Mr. LEWIS. That's just too vague to go out and ask someone to invest $100 million into precipitators and things like this in order to reduce the amount of SO_2 coming out of the stacks.

Dr. CALVERT. Well, there are various ways this could be done, obviously, and cost-effectiveness analysis as to the best ways, I think, is something that you will want to do before judgment's made on how to do it.

Let's assume there is some cost-effective method that has been demonstrated. You can try that in an area. You can try that in an area while Chris is making his measurements at his expense and you can justify whether the reception that you've got of that acid reflects the change you've made and whether the models predict that. If they do, then you've got an excuse that says now we know what's happened. We can now invest a large amount of money into now finding out what we'll need to bring back the levels to 20 kilograms per hectare per year, whatever it is that's chosen by the biologist.

Mr. LEWIS. Thank you, Dr. Calvert.

Dr. Ackermann, you also stated—and this parallels what Congressman MacKay was asking about the evidence that increased acidity is perturbing populations of microorganisms is scanty. And in the world's evolutionary process, we have seen many species vanish and others appear.

Is it possible that nature can adjust to the acid rain phenomena? It has to other changes in the environment.

Dr. ACKERMANN. Yes, that's certainly true. In our testimony, we characterize our knowledge of cause and effect as varying all the way from certain on the one hand; some of the things we know, we are quite certain of; to some things being very speculative, and the effect of the acidity of the soil on microorganisms and its long-term effect is quite speculative.

It's true that many organisms, to our discomfort, adapt themselves to our pesticides, for example, and we have to invent new ones. But on the other hand, sometimes we lose organisms. We've lost millions of different kinds of species through their environment being inhabitable. And I think that we run unknown risks when we do this kind of thing.

Mr. LEWIS. OK. Thank you. I have one last question for Dr. Bernabo.

In your testimony, you state that current data and available methods are not sufficient to establish the quantitative relationship between pollutant emissions and acid deposition on a reasonable scale.

How long do you think it will be before such methods are available, and this is a parallel to the one I asked Dr. Calvert?

Dr. BERNABO. I wish I could give you a very definitive answer on that. We are working toward, as rapidly as we can, improving the models which are available to do that. We're certain that we'll be able to do it better. How well we'll be able to do it, when, is an open question. It wouldn't be research if that weren't the case.

If we were just doing engineering and we knew we were designing something and the project would be completed at some date, but some of the best minds in the country in places like NCAR and in places—what to bear on the problem—we're using multiple approaches.

We're working with the Canadians who also have a major modeling effort in trying to develop this, and people in all parts of the world. So a major effort is underway. It's—the timing under which we'll have that, I think, is something we're not sure of.

We are looking to have—we're hoping, and again, this is planning-wise, not necessarily the reality of what will happen—breakthroughs, anything else could happen—to have something in the next 4 to 5 years that will be considerably better than the generation models we have now.

But as Dr. Calvert pointed out, there are many people in the community that are skeptical and some that are very optimistic as to how effective we'll be in reducing that type of uncertainty.

Mr. LEWIS. Well, do all three of you gentlemen feel that there's a good possibility that the relationship between Canada and the United States, that we can form a consortium in order to reduce the amount of emissions and do it in a—in a manner that's not going to, "Well, we'll test it today and if that don't work, we'll try something else tomorrow," because we're not talking about low-cost dollars; we're talking about high-cost dollars and it comes right back that—again, it's the taxpayer and consumer has to pay for this and certainly I'm not putting down the fact that we don't have

a problem with acid rain, but I feel that we got to be—all three questions—answers—have been very vague.

Dr. CALVERT. To continue the vagueness, Mr. Lewis, I hope it's not intentional, but my feeling is that if you do anything today, it's not going to be a move in the wrong direction. There are certain judgments one can make now that are realistic; that we will have an effect; that will come from our cutback of emissions. Where that effect's going to occur, we don't know.

My thought is that Chris is going to tell us where that effect occurred as we pull some effect—we're making some judgment which is built on cost-effective planning. We make that move; it's not going to break the cost of the taxpayer—break the back of the taxpayer by the high cost. We'll pick something that doesn't do that, let's hope, and we'll see whether Chris' data suggest the relationships we expect or not.

If they did not, then you're caught in a bind. What are you going to do? Obviously you're going in the right direction to cut back emissions. They do cause the acid deposition; we know that.

Mr. LEWIS. Certainly nobody's disputing that. I'm just disputing the—the lack of specificity in the data. You don't even trust it yourself.

Dr. CALVERT. I do trust it to the sense that there's going to be an effect that we'll see somewhere in eastern United States, and hopefully the models are not so bad—they'll be off by a factor of two to four in their predictions—they're not going to be so bad that we can't make value judgments——

Mr. SCHEUER. Excuse me.

Dr. CALVERT. Concerning alternatives.

Mr. SCHEUER. Will the witness yield?

What does that mean in laymen's terms, to "be off by a factor of two to four?"

Dr. CALVERT. Let's say that we predict that we want 20 kilograms per hectare per year, and that's something biologists, I think, will probably change their mind on, but that's what certainly bought by the Canadians at the moment.

The model might predict anywhere from, say, 20 to 60 in its—that's a factor of three now that we're talking about. Well, we can't miss by that much and have any faith in the model is what we're saying. The model can, though, tell differences quite well.

Mr. SCHEUER. I yield back, Tom, we have another——

Mr. LEWIS. Well, I just wanted to know if anyone else, either Dr. Ackermann or Dr. Bernabo, wanted to further comment. Otherwise, I have no further questions, Mr. Chairman.

Dr. BERNABO. I'd just like—like to comment that I think you sort of put your finger on the problem. The problem is that science is not there to give you a firm prescription with confidence limits and so forth, yet the potential threat is out there, as we've heard, and the demands of political action, and really it falls to the political arena to decide how much is society willing to risk on—in either direction—in either the possibility of inaction or the results of action.

Dr. ACKERMANN. Well, I don't feel that the uncertainties are perhaps that great. The atmospheric modeling aspect of our national program has been, to me, very disappointing because as scientists,

we would like to be able to trace the particles of air and pollutants from the stacks or wherever, the car exhausts; we'd like to trace it step by step, time and place, to the deposition and its effect and the atmospheric model was hoped it would be that—that intermediate mechanism, and it's been very disappointing. The models are not very good; they don't compare—they don't compare well with each other; we don't have very good data to correlate them with.

So—but we're persuaded that the emission data, which is the best information we have in the whole area, that the emission data is causing an effect on aquatic and terrestial plants and animals—plants particularly—irregardless of how it got there because as I did—I cited nine different converging partial indicators—no one of which might be convincing, but taken together, the area of the emissions and the area of the deposition and the timing and the kinds of material, all these things add up to us a convincing case, irregardless of the details of the mechanism in between, but nevertheless, a convincing case of cause and effect, without being able to describe the intermediate steps satisfactorily, and the intermediate steps, largely the atmospheric component, atmospheric in quantity and quality.

Mr. LEWIS. Would you say, Dr. Ackermann, that we need to spend a lot more money into our monitoring system and come up with more valid monitoring sites?

Dr. ACKERMANN. I think that our wet deposition, the rainfall, snowfall monitoring system that we will have in place this year will probably be essentially adequate for measuring the wet deposition, rain and snow.

But I think that we need to do research, followed by monitoring installations, on the dry deposition. I don't know what the size of that network is yet going to be or its cost. It will depend on the developments that are now in research, but I think we're very nearly where we need to do—need to be on rain and—rain and snow measurement.

Mr. LEWIS. If I may, Mr. Chairman, just one more question—

Mr. SCHEUER. Yes.

Mr. LEWIS [continuing]. For Dr. Calvert.

Dr. Calvert, how would we be able to measure the difference using the current monitoring sites we have right now if we spent a particular amount of money to control some of the emissions and how would we be able to see the effect in the Northeast?

Dr. CALVERT. Anything that's done in a natural system is a very difficult thing because you need a long period of time, many years of observations in order to establish a trend because there will be wet years, there'll be dry years; there'll be ones that have erratic character to them that reflects the variability of the weather.

So given a base of, let's say 5 to 10 years, one starts to get some judgment as to what the background is in that particular zone. The deposition in the New Hampshire area, we measure. The deposition in Adirondacks, we measure. One has a good quality judgment of what the present deposition is.

Starting with that base, if you make a sudden decrease in an area in the—in—let's say the Ohio and that general area where a lot of the emissions come from, Pennsylvania and New York, then it's very easy to tell whether there is a change in the deposition

that has occurred as a consequence of that cutback so that with a minor sort of perturbation on the economic scene, we can judge the quality of our control programs very quickly. We don't have to commit the whole United States to some control program which we don't know is going to work, but we know that we cut back the big emitters, there's going to be a dramatic effect somewhere and we hope it's going be in the sensitive regions.

Mr. LEWIS. Thank you, Dr. Calvert; thank you, Mr. Chairman.

Mr. SCHEUER. Buddy MacKay——

Mr. MACKAY. Thank you, Mr. Chairman——

Mr. SCHEUER [continuing]. From Florida.

Mr. MACKAY. I—I see the effort that's being made to coordinate within the United States so that the research that's done is more useful because we're all paying attention to what each other is doing.

Why would it not be equally, or even more useful to have that kind of coordination with the research that's being done in Europe and Canada, and does that coordination exist? And is not the data that they've been collecting for years useful in our studies?

Dr. BERNABO. I think the point you make is very important one. We coordinated very closely with our neighbors in Canada. In fact, we jointly sponsor a number of projects and programs. I showed some data earlier of the deposition of acid rain and it didn't stop at the U.S. border; it was a North American map that we work very closely on.

The European data, I think, as Dr. Calvert will point out with some of the stuff which he used—and his panel used in their analysis—there is no question. In the area of liming, the Swedes are spending something like $45 million a year on a liming program. We've got a lot to learn from them about the efficacy of that program and the problems and so forth.

So, there's no question, certainly at the scientific level, the individual scientist, there is a tremendous amount of coordination. We're moving very rapidly to improve that coordination in a programmatic and management level.

It is certainly to our benefit and it is certainly an integral part of solving the problem.

Mr. MACKAY. All right.

Dr. Calvert, is your conclusion about linearity and nonlinearity consistent with the Europeans' conclusions and with their data?

Dr. CALVERT. I think it's not inconsistent to speak in double negatives——

Dr. CALVERT. Yeah.

Mr. MACKAY. What does that—what does that mean?

Dr. CALVERT. Let me tell you what I really feel. The European data, of course, are obtained at a latitude which is very much further north than that in the United States. The effects are quite different. Most of the photochemistry that occurs there—it's not all photochemistry, obviously, but a lot of the action starts with the sunlight doing things to the atmosphere that causes a change.

The effects could be quite different because of that difference in latitude, because of the difference in meteorology. It's not just chemistry. There's no reason to suspect the—the Europeans are

any less intelligent or diligent or knowledgeable about acid rain than we are.

My feeling is that, indeed, their observations are real, and indeed, we have to explain them, but our first obligation is try to explain the data in the United States. At the moment, European data look to be different, and for reasons that perhaps are explicable.

Mr. MACKAY. Dr. Ackermann, I guess your group is at the highest policy level within the administration. Do you—are you satisfied that we are sufficiently coordinating our efforts, not only to take advantage of their findings, but to enable us to move as rapidly as we can from theoretical to actual practical decisions?

Dr. ACKERMANN. I think that—I'm unable to say affirmatively that we are coordinating our problematic activities as well as can be done. I would have to leave that to our Federal spokesman, but I think that we have access to all their data and to all their findings. Their research reports are of great interest over here.

I think that people have found some differences between their effects and deposition and ours, and I think, as Dr. Calvert suggested, that the—to some extent, the methods of measurement, but largely the fact that the latitude as it affects daylight and the weather systems is probably—would lead one to expect that there are these differences. So we cannot, one-for-one, take their experience and translate it here. That can't be done, but we are, I am confident, fully aware of their findings.

Mr. MACKAY. All right, thank you, sir.

Thank you, Mr. Chairman.

Mr. SCHEUER. All right.

I thank the panel. The attendance of the congressional panel that you see here is most unusual, and I think attributable to the interest that the three of you have aroused.

I must say that I was disappointed in the testimony—at least most of the testimony. I felt that the double negatives were sort of symbolic of the fuzziness, the lack of deposition, the queasiness and the faintheartedness of the testimony. The fact is that we have to make value judgments and we want to make them in a manner that is cost effective and is sensible. And we have to rely on the scientists to say, "Here is the illness and here is the remedy," or "Here is a proposed remedy."

Does the punishment fit the crime? Does the remedy—is it best calculated to meet the problem posed by the illness or is there something else that is more sensible? And you really haven't given us this kind of guidance.

We have to seek further. Maybe on some other occasion, you'll speak a little bit more directly and without so much circumlocution and backing and filling and hedging and you'll give us a little more guidance than we've been able to get out of you this morning.

In any event, we'll reconvene next Tuesday morning at 10 a.m. in this room to investigate various proposed control technologies that are used in this country and abroad and the role that they might play in an acid rain control strategy.

So until next Tuesday at 10 a.m., this hearing is adjourned.

[Whereupon, at 12:05 p.m., the subcommittees were adjourned, to reconvene at 10 a.m., Tuesday, September 20, 1983.]

ACID RAIN: IMPLICATIONS FOR FOSSIL R&D

TUESDAY, SEPTEMBER 20, 1983

HOUSE OF REPRESENTATIVES, COMMITTEE ON SCIENCE AND TECHNOLOGY, SUBCOMMITTEE ON ENERGY DEVELOPMENT AND APPLICATIONS AND THE SUBCOMMITTEE ON NATURAL RESOURCES, AGRICULTURE RESEARCH AND ENVIRONMENT,

Washington, D.C.

The subcommittees met, pursuant to recess, at 10:06 a.m., in room 2318, Rayburn House Office Building, Hon. Don Fuqua (chairman of the committee) presiding.

Present: Representatives Fuqua, Lloyd, MacKay, Nelson, Winn, Sensenbrenner, Lewis, Carney, McGrath, Boehlert, Chandler, and Gregg.

Staff present: Robert S. Kripowicz, staff director; Dr. Robert Palmer, science consultant; Dr. Harlan Watson, technical consultant; Diane Horne, detailee; Mary Beth McJury, minority technical consultant; and William Harvey, minority energy coordinator.

Mr. FUQUA. The meeting will come to order.

This is our second set of hearings on acid rain and its implications for fossil energy research and development.

In the first day of hearings in the opening remarks we discussed the three principal issues: The state of the knowledge of the causes, effects and control of acid rain; the status of current research programs and plans in these areas; and how developed and developing technologies may be used in a cost-effective manner to effect reductions which may be mandated in sulfur and nitrogen emissions.

In the hearing last week testimony was heard on the state of knowledge of the subject. While there was agreement that the vast majority of the emissions of sulfur and nitrogen in Eastern North America which cause this acid deposition are manmade, and that the reductions in emissions will reduce acid deposition, the state of knowledge is such as to give little guidance as to the best strategy to follow to mitigate the problem.

The source-receptor relationships cannot be firmly established at this time, and, due to the limited amount of data available, there is some controversy among a number of those reputable scientists in the field concerning the finding of the recent National Academy of Sciences study that changes in emissions result in proportional changes in deposition.

Today we turn to the status of the current research programs and plans in acid rain research, and how new technologies might be used to reduce emissions of sulfur and nitrogen.

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We will be hearing testimony from witnesses from the private sector and from two Government agencies primarily responsible for developing control technologies for fossil energy: the Department of Energy and the Environmental Protection Agency.

At this point I would like to insert into the record a statement by the chairman of the subcommittee that's sharing these hearings, Mr. Scheuer, and also a statement by a subcommittee chairperson, Mrs. Lloyd. We will be hearing in our first panel Dr. Mohamed El-Ashry, Director, Environmental Quality Division of TVA—he is accompanied by Mr. Michael High, the Director of the Technology Division---and also Dr. Kurt Yeager, vice president of EPRI, and Mr. William Ellison of Ellison Consultants.

If you gentlemen will please come forward.

[The opening statements of Mr. Scheuer, Mrs. Lloyd, and Mr. Winn follow:]

OPENING STATEMENT
HONORABLE JAMES H. SCHEUER
SEPTEMBER 20, 1983

THANK YOU, MR. CHAIRMAN.

TWO AND ONE-HALF YEARS AGO,
WHEN I FIRST ASSUMED THE CHAIR
OF THE SUBCOMMITTEE ON
NATURAL RESOURCES, AGRICULTURE
RESEARCH, AND ENVIRONMENT,
THE REAL ISSUE WITH RESPECT
TO ACID RAIN WAS
CONVINCING PEOPLE THAT
IT WAS IN FACT A
SERIOUS ENVIRONMENTAL THREAT.

NOW THAT MUCH, AT LEAST,
IS UNDERSTOOD.

THE ISSUE HAS BECOME: "HOW DO WE
SOLVE THIS PROBLEM IN A MANNER
THAT IS BOTH COST-EFFECTIVE
AND SENSIBLE?"

WE DO NOT WANT TO IMPOSE
CONTROL MEASURES WHICH ARE
DRACONIAN OR INEFFECTIVE.

NOR DO WE WANT TO PENALIZE UNJUSTLY
ONE REGION OF OUR COUNTRY,
LIKE THE INDUSTRIAL MIDWEST,
OR ONE SECTOR OF OUR ECONOMY,
SUCH AS THE COAL-MINING INDUSTRY.

THESE ISSUES WILL BE HOTLY DEBATED
LATER IN THIS CONGRESS, BOTH
BY THE ENERGY AND COMMERCE COMMITTEE
AND BY THE FULL HOUSE,
AS PART OF THE CLEAN AIR ACT
REAUTHORIZATION.

BY HOLDING THESE HEARINGS
LAST WEEK AND TODAY,
WE ON THE SCIENCE COMMITTEE
HAVE AN OPPORTUNITY TO MAKE
A VALUABLE CONTRIBUTION
TO THIS DEBATE.

I LOOK TO THESE HEARINGS
TO PROVIDE INFORMATION
ON A BROAD RANGE OF
CONTROL TECHNOLOGIES,
SO THAT OUR COMMITTEE
WILL BE ABLE TO OFFER ALTERNATIVES
AND TO SHAPE THE ON-GOING DEBATE
REGARDING WHICH CONTROL MEASURES
ARE COST-EFFECTIVE.

IN ORDER TO DEVISE A SENSIBLE
CONTROL STRATEGY,
WE NEED TO KNOW WHICH CONTROL STRATEGIES
WILL BE AVAILABLE,
WHAT THEY WILL COST, AND
WHEN THESE TECHNOLOGIES
WILL BE AVAILABLE.

THE QUESTION OF TIMING IS CRITICAL,
SINCE INACTION ONLY INCREASES THE DANGER
THAT OUR ENVIRONMENTAL RESOURCES
WILL BE IRREVERSIBLY DAMAGED.

BECAUSE OF THIS TREAT, IT IS CLEAR
THAT NOW IS THE TIME
TO END THE STALEMATE
IN THE ACID RAIN DEBATE,
BOTH BY STEPPING UP OUR USE
OF THOSE CONTROL TECHNOLOGIES
WHICH ARE CURRENTLY THE MOST COST-EFFECTIVE
AND BY DESIGNING
A SENSIBLE COMPREHENSIVE CONTROL STRATEGY
WHICH WILL MINIMIZE THE
LONG-TERM IMPACTS OF ACID RAIN.

MR. CHAIRMAN, LET ME CLOSE BY SAYING
THAT WE HAD HOPED TO HAVE
EPA ADMINISTRATOR BILL RUCKELSHAUS
WITH US TODAY TO DESCRIBE
THE STRATEGY THAT EPA IS EVOLVING
FOR DEALING WITH ACID RAIN.

UNFORTUNATELY, THE ADMINISTRATION
HAS NOT YET REACHED A CONSENSUS
ON ITS RESPONSE.

I AM SURE THAT YOU WOULD JOIN WITH ME
IN EXTENDING TO BILL RUCKELSHAUS
AN OPEN INVITATION
TO USE OUR SUBCOMMITTEES
AS A PUBLIC FORUM
WHEN HE IS PREPARED
TO DELINEATE THE ADMINISTRATION'S
POLICY ON ACID RAIN.

THANK YOU, MR. CHAIRMAN.

OPENING STATEMENT OF THE
HON. MARILYN LLOYD

ACID RAIN HEARING

SEPTEMBER 20, 1983

THANK YOU, MR. CHAIRMAN. I AM VERY PLEASED TO BE ABLE TO PARTICIPATE IN TODAY'S HEARING ON ACID RAIN. I KNOW THAT THIS HEARING AND THE ONE LAST WEEK WILL BE IMPORTANT, NOT ONLY DURING OUR CONSIDERATION OF THE REAUTHORIZATION OF THE CLEAN AIR ACT, BUT TO THE FUTURE OF THE COAL INDUSTRY AND ELECTRIC UTILITIES OF THIS COUNTRY AS WELL.

I WANTED TO MENTION FOR THE RECORD A LETTER WHICH WAS SIGNED BY CHAIRMAN FUQUA, CHAIRMAN SCHEUER AND MYSELF, AND WAS SENT TO THE NATIONAL LABORATORY CONSORTIUM OF THE INTERAGENCY TASK FORCE ON ACID DEPOSITION TO ASK FOR THEIR EVALUATION OF THE RECENTLY PUBLISHED NATIONAL RESEARCH COUNCIL REPORT ON ACID DEPOSITION IN EASTERN NORTH AMERICA.

THE CONSORTIUM RESPONSE TO US INDICATED THAT THERE ARE STILL MANY SIGNIFICANT DISAGREEMENTS WITHIN THE SCIENTIFIC COMMUNITY IN THE AREAS OF VERIFYING LINEARITY AND THE STATE AND ACCEPTABILITY OF CURRENT MODELS. I MENTION THESE AS ONLY TWO EXAMPLES OF AREAS OF WIDE UNCERTAINTY WHICH EXIST IN OUR EFFORTS TO DETERMINE THE CAUSES AND EFFECTS OF ACID RAIN.

I FEEL THAT IT IS MOST IMPORTANT THAT WE RECOGNIZE THESE DIFFERENCES OF OPINION AND CAREFULLY EVALUATE AND SELECT THE MOST PRUDENT RESEARCH PROGRAM AVAILABLE TO US BEFORE WE MAKE REGULATORY DECISIONS WHICH MAY HAVE LONG-TERM IMPACTS NOT ONLY ON THE ENVIRONMENTAL BUT ALSO ON THE ECONOMIC WELL-BEING OF OUR NATION.

IN CLOSING, MR. CHAIRMAN, I WOULD LIKE TO TAKE A MOMENT TO WELCOME OUR TWO REPRESENTATIVES FROM TVA, MR. MOHAMED EL-ASHRY AND MR. MICHAEL HIGH. I THINK WE ARE ALL AWARE OF THE CONTRIBUTIONS THAT THE TVA HAS MADE IN OUR COUNTRY'S ENERGY DEVELOPMENT, NOT ONLY IN MY OWN AREA OF TENNESSEE BUT THROUGHOUT THE SOUTHEASTERN UNITED STATES. GENTLEMEN, WE APPRECIATE THE REMARKABLE WORK THAT HAS BEEN DONE BY TVA AND WE LOOK FORWARD TO YOUR TESTIMONY.

THANK YOU, MR. CHAIRMAN.

OPENING STATEMENT BY THE HONORABLE LARRY WINN, JR.
ACID RAIN HEARING: IMPLICATIONS FOR FOSSIL ENERGY R&D
SEPTEMBER 20, 1983

THANK YOU, MR. CHAIRMAN, FOR THIS OPPORTUNITY TO COMMENT ON THESE HEARINGS AND TO WELCOME OUR WITNESSES TODAY.

IN OUR FIRST HEARING ON THE ACID RAIN SUBJECT LAST WEEK, WE RECEIVED ILLUMINATING INFORMATION ON THE SUBJECT. WE WERE ADVISED THAT THE STUDY OF ACID RAIN HAS BEEN PURSUED FOR A RELATIVELY SHORT PERIOD OF TIME, THAT DATA ARE BEING ACCUMULATED AT A GRATIFYING RATE, THAT THERE ARE A NUMBER OF QUESTIONS BEING ANSWERED, THAT THERE ARE EVEN MORE QUESTIONS BEING RAISED, AND THAT THE SUBJECT PROBABLY WILL NOT BE DEFINED FOR AN EXTENDED PERIOD. WE HEARD THAT IT MAY BE AS MUCH AS 20 YEARS BEFORE WE HAVE ACID RAIN IN PROPER PERSPECTIVE.

THIS LEADS US TO A SERIOUS CONFRONTATION. AS YOU KNOW, MR. CHAIRMAN, THERE ARE A NUMBER OF PEOPLE WHO WOULD REQUIRE IMMEDIATE INSTALLATION OF LARGE AND EXPENSIVE FACILITIES TO REDUCE THE AMOUNT OF SULFUR AND NITROGEN EMISSIONS FROM FOSSIL BURNING PLANTS. IN VIEW OF THE MEAGER AMOUNT OF INFORMATION WE NOW POSSESS, IT WOULD APPEAR THAT PRESENTLY-PROPOSED ACID RAIN LEGISLATION REQUESTS TOO MUCH, TOO SOON.

I CERTAINLY THINK THAT INTELLIGENT CORRECTIVE ACTION CAN TAKE PLACE BEFORE TOTAL KNOWLEDGE IS ACQUIRED ON ACID RAIN OR ANY OTHER POSSIBLE PROBLEM. HOWEVER, I REITERATE THAT BEFORE WE TAKE EXPENSIVE,

TIME-CONSUMING, AND MIS-DIRECTED STEPS TO CORRECT A PROBLEM, WE HAD FIRST BETTER DEFINE THE PROBLEM AND THEN WORK TOWARD ALLEVIATING THAT PROBLEM.

I NOTICED THAT OUR WITNESSES TODAY INCLUDE A WIDE RANGE OF INTERESTS. THE TENNESSEE VALLEY AUTHORITY BURNS LARGE QUANTITIES OF FOSSIL FUELS IN GENERATING ELECTRICITY THROUGHOUT THAT AREA OF THE UNITED STATES. THEY CERTAINLY ARE INTERESTED IN DOING SO EFFICIENTLY AND IN AN ENVIRONMENTALLY ACCEPTABLE MANNER. THE ELECTRIC POWER RESEARCH INSTITUTE CONDUCTS RESEARCH FOR THE VAST MAJORITY OF UTILITY COMPANIES IN THE UNITED STATES. THEY ALSO ARE INTERESTED IN THE ENVIRONMENTAL ASPECTS OF POWER GENERATION. BOTH THE ENVIRONMENTAL PROTECTION AGENCY AND THE DEPARTMENT OF ENERGY ARE WORKING TO ALLEVIATE HARMFUL EMISSIONS. THE KNOWLEDGE THAT THEY AND MR. ELLISON ACCUMULATE ON REDUCTION OF EXISTING STATIONARY EMISSIONS WILL BE BENEFICIAL WHEN THE TIME COMES TO MAKE APPROPRIATE CHANGES IN THOSE EMISSIONS, WHICH WOULD BE DIRECTED FROM DATA ACCUMULATED ON ACID RAIN CAUSES AND AFFECTS.

MR. CHAIRMAN, IT IS HOPED THAT THESE HEARINGS WILL PROVIDE AN OBJECTIVE RECORD ON ACID RAIN. WITH SUCH INFORMATION IN HAND, WE CAN ADDRESS THOSE AREAS FOR WHICH WE DO NOT HAVE COMPLETE INFORMATION. IN THAT WAY WE CAN THEN CLEANUP THE PROBLEMS WHICH ARE IDENTIFIED AND DEFINED.

THANK YOU, MR. CHAIRMAN.

Mr. FUQUA. I might say as a custom of the committee, if you wish to insert your prepared statement for the record, we'll do that, and if you wish to summarize, that would certainly be permissible.

Dr. El-Ashry?

STATEMENT OF DR. MOHAMED T. EL-ASHRY, DIRECTOR, ENVIRONMENTAL QUALITY DIVISION, TENNESSEE VALLEY AUTHORITY, ACCOMPANIED BY DR. MICHAEL D. HIGH, DIRECTOR, ENERGY DEMONSTRATIONS AND TECHNOLOGY DIVISION, TENNESSEE VALLEY AUTHORITY

Dr. EL-ASHRY. That's exactly what we are planning on doing.

Mr. FUQUA. Thank you.

Dr. EL-ASHRY. Insert the statement for the record. Then we'll summarize in about 12 minutes or so.

Mr. FUQUA. OK.

Dr. EL-ASHRY. Mr. Chairman, we are pleased to be here today in response to your invitation to present information on acid rain. We should emphasize, however, that TVA's conclusions and its recommendations as set forth in this testimony are not those of the administration. The TVA Board of Directors feels that TVA, as a Federal corporation as well as the Nation's largest electric system, has the obligation to share its experiences and perspective with those in the legislative and executive branches who are grappling with this issue.

As a result of TVA's experience in solving interrelated energy and environmental problems, TVA has knowledge in two areas which are highly relevant to the acid rain problem.

One, having reduced the annual sulfur dioxide emissions from its coal-fired powerplants by about a million tons since 1978, TVA has considerable knowledge about the problems and the costs of pollution control technology. Two, TVA has been monitoring the acidity of precipitation in the Tennessee Valley since 1971. For the past 5 years, TVA has also been engaged in research on the atmospheric transport and transformation processes as well as on the effects of acid deposition on the region's forests, soils, water quality, and fisheries.

Based on this experience, TVA believes control technologies currently available to reduce sulfur emissions to the atmosphere can be applied at reasonable costs and that these controls should be applied now.

Precipitation data since 1971 in the Tennessee Valley Region indicates pH's ranging from 4.1 to 4.4 on an average annual basis, which are comparable to values observed for New England rainfall. In addition, there have been reports of a trend toward decreased levels of pH and increases in the aerial extent of low pH rain water, particularly toward the Southeast, and these are of particular concern to us.

We have no direct evidence of a link between acid deposition and impacts to the resources in the Tennessee Valley. However, detailed investigations of several recent observations in the Tennessee Valley region may reveal that some observed impacts could very well be related to acid deposition. For example:

One, pH levels in some streams are depressed after rainfalls to an extent that aquatic life may be impacted.

Two, rainbow trout mortalities have occurred in two trout rearing facilities following heavy rainfalls. Recent studies by TVA indicate that the stream acidity and the resulting increases in stream aluminum concentration contributed to these kills. Acid soils in the watershed appeared to be a major contributor to low stream pH. But, the contribution of acid deposition to acidification in the watershed could not be precluded and is being investigated further.

Three, an unusually high incidence of skeletal deformities in smallmouth bass has been reported recently from Chatuge Reservoir.

Four, preliminary investigations indicate mortality, decreased growth rates, and buildup of metals in plant tissues of some tree species in the Tennessee Valley region.

TVA is interested in these effects, and we are now assessing the extent of damages and their causes. TVA recognizes, however, that it may take many years before we can determine and quantify the role acid deposition may be having in these effects. In fact, we may never be able to fully understand the complex processes by which these effects occur. Nonetheless, the environmental impacts of acid deposition may be irreversible and sensitive ecosystems may presently be at risk. Accordingly, TVA supports further reductions in SO_2 and NO_x emissions.

We view acid deposition as a local, regional, national, and international problem that's not adequately addressed by present requirements of the Clean Air Act.

An important contribution TVA has made as one of the Nation's largest consumers of coal is to invest the necessary funds to cut its SO_2 emissions in about half, from about 2 million to 1 million tons per year. This is an important contribution both in fact and by way of example to reduce the total burden of sulfur pollution. This emission control effort involved a capital cost of $724 million with annualized cost of $334 million, representing an increased cost to TVA ratepayers of 6 to 8 percent.

The program undertaken consists of building scrubbers, coal washing plants, and switching to lower sulfur coal.

Pollution control comes with a price, and TVA and its power customers are paying that price. However, we accept these costs as an expense of producing electricity from coal and consider the costs within the limits of practicality.

A much less expensive way to limit total emissions is to invest in energy conservation measures. TVA's experience in the last 6 years has shown that we would be saving more than 5.7 billion kilowatts annually by 1990, and we expect these efforts to reduce generating capacity requirements by more than 3,000 megawatts. If TVA had to supply 5.7 billion kilowatt hours of power from one of its existing coal-fired plants, it could mean an additional burden of sulfur dioxide emissions of about 90,000 tons per year.

TVA's pollution control experience demonstrates that sulfur dioxide emissions can be reduced at reasonable costs. However, it should be recognized that TVA's results cannot be applied casually to all utility systems since the mix of energy sources affects the

flexibility available to a utility to keep costs of emission control low.

The seriousness of today's acid deposition problem indicates that TVA may need to achieve further reductions in its total SO_2 emissions in the future. TVA has the ability and willingness to further reduce its total SO_2 emissions if a national reduction goal, including TVA's fair share of that goal, is established.

We believe that an equitable control mechanism would require emission reductions for the 31 acid deposition control States to be proportional to each State's share of total SO_2 emissions for the 1980 base year. States should have the flexibility to allocate their emission reduction to any or all sources. Emissions trading and offsets for utility systems and other sources, both within and between States, should be allowed by the States.

After the States determine the SO_2 emission reductions that are required from each source, multiple sources such as utility systems should be free to choose the most cost-effective approach to obtain their allocated reductions regardless of State lines. For example, systems should be given credit for reductions achieved through conservation efforts, reduced plant usage, or shifts to other generation methods.

Using this approach, and if permitted to use the most cost-effective emission reduction techniques, TVA can meet its fair share of an 8-million-ton national reduction, assuming loads would remain about where they are now, at no cost whatsoever to its consumers. In comparison, a national goal of 12 million tons would cost TVA's ratepayers somewhere between 4 and 6 percent a year.

In summary, TVA recognizes that environmental impacts of acid deposition may be irreversible and that sensitive ecosystems may presently be at risk, and we believe that actions to further control SO_2 and NO_x emissions are warranted at this time.

The availability and abundance of coal make it an important energy option. The environmental effects of increased coal combustion, however, stand as a principal deterrent to its use. We believe that the key to increased reliance on coal lies in minimizing those environmental effects by continued improvements in both control and combustion technologies. Accordingly, the Nation should maintain a high priority for research and demonstration of innovative technologies for the reduction of emissions from coal, including atmospheric fluidized bed combustion.

At this point, Mr. Chairman, I would like to defer to my colleague, Dr. High, who will discuss TVA's experience in the research and demonstration of emission control technology.

Thank you.

Dr. HIGH. TVA has done a considerable amount of pioneering work in the SO_2 emission control technology since the late 1960's, primarily in atmospheric fluid bed combustion, coal cleaning, and flue gas desulfurization. Such research and demonstration not only improve our ability to control emissions, but also lead to cost savings by allowing economically and environmentally acceptable use of the region's abundant high sulfur coal resources.

To achieve rapid commercial acceptance by the electric power industry of the fluidized bed combustion, technology needs a utility-scale demonstration to verify reliability, performance, and costs.

TVA, with significant support from Congress, has taken the first step toward that utility-scale demonstration by constructing a $68 million, 20-megawatt pilot plant in Paducah, Ky. Construction on this activity is complete, and the unit was shaken down and operations began in May 1982. To date, that pilot plant has operated some 3,250 hours and consumed approximately 18,000 tons of coal and 8,000 tons of limestone.

A major test program, funded jointly by the Electric Power Research Institute and ourselves, is underway to develop that technology and environmental performance data base for the design and construction of a demonstration plant.

Recent studies have convinced us that the next major step is a utility-scale demonstration facility. This can be accomplished, we think, by replacing an existing pulverized, coal-fired boiler with an AFBC boiler. This approach would accomplish the goals of demonstrating the essential elements of the technology at a significantly lower cost than a stand-alone facility.

We propose such a facility to be located at our TVA coal-fired Shawnee plant in Paducah, Ky. The design fuel of such a demonstration would be high-sulfur, Illinois-basin coal, such as that typified by Kentucky No. 9 coal seam. Fuel flexibility, however, of this demonstration facility would be recognized in a test program. We estimate the preliminary cost estimate for construction of this facility to be $200 million, with about a $20 million test program in addition to that. We and EPRI, Duke Power Co., and the State of Kentucky, have, in fact, affirmed our commitment to this project. We committed some $135 million, to date, to that.

TVA's projections indicate that a new AFBC boiler fueled with Illinois-basin, high-sulfur coal would produce electricity at a cost of some 10 to 20 percent less than the conventional plant with a scrubber, and such a plant could achieve greater than 90-percent sulfur removal and produce less oxides of nitrogen than a conventional plant.

Based on our present schedule, the atmospheric fluidized bed combustion we think can be fully commercial by mid-1990.

In the area of coal cleaning, TVA has cleaned approximately 18 million tons of our 27.5 million ton coal burn in 1982. Improvements in coal cleaning offer many advantages to TVA. Our program focuses on process improvements, process control, and resource recovery in the coal cleaning area.

We are demonstrating, at our Paradise steam plant, high-gradient magnetic separation technology which improves Btu recovery and sulfur reduction in the fine coal washing circuit. We are also conducting research on oil agglomeration and magnetite recovery processes which recover clean coal from the fine refuse and extract magnetite from that coal ash.

A major program in cooperation with the Electric Power Institute involves a demonstration of prompt neutron activation analysis techniques, such as the rapid sulfur meter and continuous online nuclear analyzer of coal, to monitor and control coal wash production plant operation. Testing of the rapid sulfur meter and the CONAC, as it's called, is now underway at our Paradise plant.

Additional research is needed in the coal cleaning area to investigate cost-effective chemical coal cleaning technology that may be

tailored to remove almost any desired amount of sulfur from coal, especially for those cases where greater than 50 to 60 percent reduction in sulfur is needed.

When applicable as a compliance strategy, coal cleaning is in most cases the least costly control technology option. Because of the variability of coal quality, the cost and the amount of sulfur removal vary widely. The overall sulfur content as well as the form and distribution of the sulfur greatly influence the cost of coal cleaning.

For example, for U.S. mid-bituminous eastern coals physical coal cleaning techniques are capable of removing up to 35 percent of the coal-sulfur in the coal when the content is 3 to 5 percent. Coal cleaning we think adds somewhere between 15 and 20 percent to the cost of electric power generation.

In the area of flue-gas desulfurization, TVA's major role began in the development efforts in 1972 with a cooperative effort with the Environmental Protection Agency for construction of three 10-megawatt [sic] pilot plant scrubber facilities, again at our Shawnee steam plant in Paducah, Ky. Since then, we've spent more than $35 million by the EPA, EPRI, and ourselves, and the Department of Energy at this facility developing wet limestone scrubbers. Numerous technology advances are available today because of this effort.

For example, developments in forced oxidation and waste disposal have enabled us to retrofit these technologies into our full-scale scrubbers at our Widows Creek 7 and 8 units. We are also very active with EPA in preparing technology assessments and cost estimates for both recovery and throwaway processes for flue-gas desulfurization.

Today our efforts primarily are focused on demonstrating dry FGD technology, commonly known as spray drayer. We have two such facilities—experimental facilities—at our Shawnee unit. These two plants are supplied by different vendors and providing critical data necessary to adopt this technology in the Eastern United States.

We think other technologies that need further development or demonstration include double alkali using limestone, limestone injection multiple burners, dry FGD with limestone, lower cost recovery systems, and those retrofittable technologies for existing outdated limestone scrubbers, combined with NO_x and SO_2 reductions.

I believe this subcommittee will hear more about these technologies from testimony by EPRI and EPA today.

FGD is capable of removing 90 percent of the sulfur from utility boiler flue gas. Although there are problems associated with operating and maintaining scrubbers as a retrofit, this technology may be the choice in the near term. Costs associated with FGD we think will add approximately 50 percent to the cost of electricity at coal-fired powerplants.

Mr. Chairman, this summarizes very quickly some of TVA's activities in the control technologies.

Thank you.

[The prepared statement of the Tennessee Valley Authority follows:]

ACID RAIN STATEMENT OF
THE TENNESSEE VALLEY AUTHORITY
BEFORE THE SUBCOMMITTEES ON ENERGY DEVELOPMENT AND APPLICATION AND
NATURAL RESOURCES, AGRICULTURE RESEARCH AND ENVIRONMENT
U.S. HOUSE OF REPRESENTATIVES
WASHINGTON, D.C., SEPTEMBER 20, 1983

We are pleased to be here today in response to your invitation to present information on acid rain. Our testimony is based upon knowledge about acid rain issues in the areas requested by the Subcommittees. We should emphasize that TVA's conclusions and its recommendations as set forth in this testimony are not those of the Administration. As you know, this issue is currently under review by the Administration. The TVA Board of Directors feels that TVA, as a federal corporation as well as the nation's largest electric system, has the obligation to share its experiences and perspective with those persons in the legislative and executive branches who are grappling with this issue.

As the Nation's largest producer of electricity as well as a Federal instrumentality providing for the stewardship of the Tennesse Valley region's natural resources, TVA is well qualified to be of assistance to the Subcommittees and to the Congress. As a result of TVA's experience in solving interrelated energy and environmental problems, TVA has knowledge in two areas which are highly relevant to acid rain:

- Having reduced the annual sulfur dioxide emissions from TVA coal-fired generating plants by about a million tons since 1978, TVA has considerable knowledge about the problems and the costs of pollution control technology; and
- TVA has been monitoring the acidity of precipitation in the Tennessee Valley since 1971. For the past five years, TVA has also been engaged in research on the atmospheric transport and transformation processes

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as well as on the effects of acid deposition on the region's forests, soils, water quality, and fisheries.

Based on this experience, we first present a summary of TVA's knowledge regarding the impacts of acid deposition on the Tennessee Valley region and the possible causes of the Valley's rainfall acidity. We then summarize our experience in controlling emissions. Finally, we discuss our program to develop and demonstrate improved technologies for further controlling sulfur dioxide emissions. As a result of this experience, TVA believes control technologies currently available to reduce sulfur emissions to the atmosphere can be applied at reasonable costs and that these controls should be applied.

Impacts of Acid Rain in the Tennessee Valley Region

Acid deposition is one of the most significant environmental issues of this decade. The fact that rainfall acidity in this region is as high as that for any region in the eastern United States is a matter in which TVA is vitally interested. Although there is little direct evidence of acid deposition impacts, the circumstantial evidence currently available has added to the belief that acid deposition may result in long-term damage to our sensitive ecosystems.

TVA began monitoring the acidity of precipitation at two stations in 1971. In 1978 TVA began a regional (17 stations) deposition monitoring network. These data indicate pH's ranging from 4.1 to 4.4 on an average annual basis (measured pH levels range from 3.7 to 6.5)--typical of the values for New England rainfall. We have also found that the pH of rainfall at Tennessee Valley sites remote from TVA power plants is similar to that at monitoring sites at our power plants. We have not been able to detect any

trend of increasing or decreasing acidity. Therefore we cannot confirm from our monitoring data the apparent trend exhibited by comparison of data on the pH of rainfall over the entire eastern United States over an 18-year period between 1955 and 1973. These comparative data indicated a trend toward decreased levels of pH and increases in the aerial extent of low pH rainwater and are of particular concern to us because most of the increase seems to be toward the Southeast.

We have no direct evidence of a link between acid deposition and impacts to the resources in the Tennessee Valley. However, detailed investigations of several recent observations in the Tennessee Valley region may reveal that some observed impacts could be related to acid deposition:

- o pH levels in some streams are depressed after rainfalls to an extent that aquatic life may be impacted.
- o Rainbow trout mortalities have occurred in two trout rearing facilities following heavy rainfalls. Recent studies by TVA indicate that the stream acidity and resulting increases in stream aluminum concentration contributed to these kills. Acid soils in the watershed appeared to be a major contributor to low stream pH. But, the contribution of acid deposition to acidification in the watershed could not be precluded and is being investigated further.
- o An unusually high incidence of skeletal deformities in smallmouth bass has been reported recently for one headwater reservoir (Chatuge Reservoir). However, other factors such as pesticides and herbicides in watershed runoff may be responsible for this.
- o Analysis of comparative data on sulfur cycling in watersheds near and distant from a coal-fired power plant indicate that there may be more of a buildup of sulfur at the site near the power plant.

- Preliminary investigations indicate mortality, decreased growth rates, and buildup of metals in plant tissue of some tree species in the Tennessee Valley region. Of course, many factors such as drought, disease, or other environmental pollutants could be responsible for such changes; but the possibility exists that acid deposition may be contributing to a loss in forest productivity.

TVA is interested in these effects and we are now assessing the extent of damages and their causes. Our initial focus in these assessments is on investigations of the effects of acidic inflows on the region's reservoirs, the influence of soil chemical and physical properties on surface water quality in forested watersheds, causes of deformities in smallmouth bass in high elevation reservoirs, and trends in forest productivity in high elevation watersheds. We are also continuing our program to monitor rainfall quality and to develop improved techniques for predicting the transport and transformation of air pollutants. TVA recognizes, however, that it may take many years before we can determine and quantify the role acid deposition may be having in these effects. In fact, we may never be able to fully understand the complex processses by which these effects occur. None theless, the environmental impacts of acid deposition may be essentially irreversible and sensitive ecosystems may presently be at risk. Accordingly, TVA supports further reductions in SO_2 and NO_x emissions.

Suspected Causes of Acid Deposition

TVA recognizes the likelihood of a relationship between acid deposition and the total load of SO_2 and NO_x in the atmosphere. TVA also recognizes that long-range transport and transformation of pollutants in the

plumes of fossil-fueled boilers of all types produce sulfate and nitrate particles which are believed to be linked to acid deposition. TVA views acid deposition as a local, regional, national, and international problem that is not adequately addressed by present requirements of the Clean Air Act and supports further reductions in SO_2 and NO_x emissions.

Experience in Controlling Emissions

An important contribution TVA has made as one of the Nation's largest consumers of coal is to invest the necessary funds to cut its SO_2 emissions in about half from about two to one million tons per year. This pollution control program undertaken to comply with the Clean Air Act combined building scrubbers, coal washing plants, and switching to lower sulfur coal. This is an important contribution, both in fact and by way of example, to reduce the total burden of sulfur pollution. This emission control effort involved a capital cost of $724 million with annualized cost of $334 million--representing an increased cost to TVA ratepayers of 6-8 percent.

Pollution control comes with a price, and TVA and its power customers are paying the price for complying with the Clean Air Act. However, TVA accepts these costs as an expense of producing electrical energy from coal and considers the costs within the limits of practicality.

A much less expensive way to limit total emissions is to invest in energy conservation measures. Conservation by now is recognized as the cleanest source of energy, but achieving results requires more than lip service. TVA's experience in the last six years has shown that education, financial incentives, and state-of-the-art conservation technology can be combined into an overall effort that achieves sizable results. By 1990 we

expect our conservation efforts to be saving more than 5.7 billion kWh annually, and we expect these efforts to reduce generating capacity requirements by more than 3,000 MW. If TVA had to supply 5.7 billion kWh of power from one of its existing coal-fired plants, it could mean an additional burden of sulfur dioxide emissions of about 90,000 tons per year.

Future Control Capabilities and Associated Costs

TVA's pollution control experience demonstrates that sulfur dioxide emissions can be reduced at reasonable costs. However, it should be recognized that TVA's results cannot be applied casually to all utility systems since the mix of energy sources affects the flexibility available to a utility to keep costs of emission control low. The seriousness of today's acid deposition problem indicates that TVA may need to achieve further reductions in its total SO_2 emissions in the future. TVA has the ability and willingness to further reduce its total SO_2 emissions if a national reduction goal--including TVA's fair share of that goal--is established. TVA estimates that to achieve its share of an 8-million-ton per year national reduction in sulfur dioxide emissions would require TVA to further reduce its SO_2 emissions to about 0.7 million tons per year and would increase the rates for TVA electricity by about 3 to 6 percent depending on future TVA system load demands. A 10-million-ton per year national reduction would reduce TVA's SO_2 emissions to about 0.5 million tons per year increasing rates by 6 to 8 percent. These estimates assume that each State's share of the national reduction is based on utility SO_2 emissions only and that offsets for new SO_2 sources are not required.

TVA believes that a fairer allocation would require emission reductions for the 31 acid deposition control States to be proportional to

each State's share of total SO_2 emissions for the 1980 base year. States should have the flexibility to allocate their emission reduction to any or all sources. Emissions trading and offsets for utility systems and other sources both within and between States should be allowed by the States. In determining a State's compliance in achieving its share of emission reductions, credit should be given to reflect trading between sources in different States. NO_x and SO_2 emission reduction trading should be allowed on a two-for-one basis.

After the States determine the SO_2 emission reductions that are required from each source, multiple sources such as utility systems should be free to choose the most cost effective approach to obtain their allocated reductions regardless of State lines. For example, systems should be given credit for reductions achieved through conservation efforts, reduced plant usage, or shifts to other generation methods.

Using this approach and if permitted to use the most cost-effective emission reduction techniques--such as emissions reduction trading within and between States, shifts to other generation methods, and conservation efforts--TVA can meet its fair share of a 12-million-ton national reduction goal with an increase in electric rates of only 4 to 6 percent. The total annual cost to TVA would be between $160 million and $340 million (depending on future load demands) and total SO_2 emissions from the TVA system would be reduced to about 0.6 million tons per year. By comparison, TVA can meet its fair share of an 8-million-ton national reduction using this approach at a total annual cost of $0 to $150 million (depending on future load demands) under TVA's preferred alternative scheme. We are attaching with this statement a table outlining the cost to TVA of various SO_2 reductions. (Attachment 1).

TVA would like to stress that conservation should be recognized as a least-cost method of achieving sulfur dioxide emission reductions. Many utilities' estimates of the cost of compliance do not reflect this fact which TVA has demonstrated. Intensive industry wide conservation efforts could reduce power requirements and corresponding SO_2 emissions by a considerable amount. With a strong conservation effort alone, TVA has the potential of reducing its annual power output by up to 20 billion kWh by the year 2000. Utilities should be expected to finance conservation, and estimates of compliance cost should reflect conservation opportunities.

Emission Control Technology Research and Demonstration

TVA has done pioneering work in SO_2 emission control technology since the late 1960s--primarily in atmospheric fluidized bed combustion, coal cleaning, and flue gas desulfurization. Such research and demonstration not only improve our ability to control emissions but also lead to cost savings by allowing economically and environmentally acceptable use of the region's abundant high-sulfur fuels resources.

Atmospheric Fluidized Bed Combustion (AFBC)--To achieve rapid commercial acceptance by the electric power industry, this technology needs a utility-scale demonstration to verify reliability, performance, and costs. TVA, with significant support from Congress, has taken the first major step leading to a utility-scale demonstration by constructing a $68-million, 20-MW AFBC pilot plant in Paducah, Kentucky. Construction activities on this facility are complete, and the unit began shakedown operations in May 1982. The pilot plant has operated approximately 3,250 hours and consumed approximately 18,246 tons of coal and 8,482 tons of limestone. A major test program, funded jointly by the Electric Power Research Institute

and TVA, is underway to develop the technical and environmental performance data to be used in the design and construction of a demonstration plant.

Based on recent studies, TVA has concluded that the next major step, a utility-scale demonstration facility, can be accomplished by replacing an existing pulverized, coal-fired boiler with an AFBC boiler. This approach will accomplish the goals of demonstrating the essential elements of the technology at a significantly lower cost than a new standalone facility.

The AFBC demonstration facility would be located at TVA's coal-fired Shawnee Steam Plant in Paducah. To demonstrate the technology, an AFBC boiler would be constructed beside an existing unit at the plant as a replacement for the existing boiler. The AFBC boiler would be capable of generating approximately 160 MW of electric power and would make maximum use of the existing turbogenerator set and other associated equipment. The design fuel for the demonstration would be high-sulfur, Illinois-basin coal, as typified by Kentucky No. 9 seam coal. However, fuel flexibility will be considered by testing alternate fuels during the test program. The preliminary cost estimate for construction of the AFBC demonstration at Shawnee is $200 million. It is anticipated that a 3-year test and demonstration period will cost approximately $20 million. EPRI, the Commonwealth of Kentucky, and Duke Power are cooperating with TVA on this project.

TVA projections indicate that a new AFBC boiler fueled with Illinois-basin, high-sulfur coal will produce electricity at a cost 10 to 20 percent less than a conventional plant with scrubber. Such a plant could achieve greater than 90 percent sulfur removal and would produce less oxides of nitrogen than a conventional plant.

Coal Cleaning--Of the 27.5 million tons of coal burned by TVA in 1982, approximately 18 million tons were washed either at the mine site or at a TVA-operated facility. Improvements in cleaning coal offer many advantages to TVA. Our program focuses on process improvements, process control, and resource recovery.

We are demonstrating at our Paradise Steam Plant high gradient magnetic separation technology which improves Btu recovery and sulfur reduction in the fine coal washing circuit. We are conducting research on oil agglomeration and magnetite recovery processes which recover clean coal from fine coal refuse and extract magnetite from fly ash respectively. A major program involves the demonstration of prompt neutron activation analysis techniques, rapid sulfur meter (RSM) and continuous online nuclear analyzer of coal (CONAC), to monitor and control coal wash plant operation. Testing of RSM and CONAC is now underway at Paradise.

Additional research is needed to develop some cost-effective chemical coal cleaning technology that may be tailored to remove almost any desired amount of sulfur from coal, especially for those cases where greater than 50- to 60-percent reduction in sulfur is required.

For CONAC, in addition to its use in the quality control and optimization of coal cleaning operations, we will investigate its applications to real time best rate control, optimization of power system load dispatch, and boiler slagging and fouling control.

When applicable as a compliance strategy, coal cleaning in most cases is the least costly control technology option. Because of the variability of coal quality, the cost and the amount of sulfur removal vary widely. The overall sulfur content as well as the form and distribution of the sulfur greatly influence the cost of coal cleaning. For U.S.

mid-bitiuminous eastern coals physical coal cleaning techniques are capable of removing up to 35 percent of the sulfur in the coal. With sulfur contents in the range of 3 to 5 percent, physical coal cleaning adds 15 to 20 percent to the cost of electric generation.

Flue Gas Desulfurization (FGD)--TVA's major role in FGD development began in 1972 with the construction of three 10-MW pilot plants at our Shawnee Steam Plant in Paducah, Kentucky. Since then, more than $35 million has been invested by the Environmental Protection Agency, EPRI, the Department of Energy, TVA, and others at this facility. Numerous technology advancements are available today because of this effort. For example, developments in forced oxidation and waste disposal have enabled us to retrofit these technologies into the full-scale scrubbers at our Widows Creek units 7 and 8. We are also active in preparing technology assessments and cost estimates for both recovery and throwaway processes. These estimates are quoted throughout the industry.

Today, our efforts are focused on demonstrating dry FGD technology (commonly known as spray dryer) on high-sulfur coal at Shawnee. Two pilot plants supplied by different vendors are providing critical data necessary to adopt this technology in the eastern United States.

Based on recent studies (cost estimates and pilot plant results) and major cost sharing by a vendor, it is highly likely that TVA will demonstrate dry FGD technology on a 160-MW, high-sulfur, coal-fired unit at Shawnee. The resolution of engineering uncertainties at this level is a necessary prerequisite to full adoption across the industry.

Other technologies that need further development or demonstration include double alkali using limestone; limestone injection multiple burner;

dry FGD with limestone; lower cost recovery systems; retrofittable technologies for existing outdated limestone scrubbers; and combined NO_x/SO_2 removal processes.

FGD is capable of removing over 90 percent of the sulfur from utility boiler flue gas. Although there are problems associated with operating and maintaining scrubbers as a retrofit, FGD may be the technology of choice. Costs associated with FGD add 50 percent to the cost of electricity at coal-fired power plants.

SUMMARY

TVA recognizes that environmental impacts of acid deposition may be essentially irreversible and that sensitive ecosystems may presently be at risk. The acidity of rainfall in the Tennessee Valley and some recently observed effects that may be caused by acid deposition are of serious interest to TVA. Although we are investigating the extent of damage and the causes of recognized problems, TVA believes that actions to further control SO_2 and NO_x emissions are warranted at this time.

TVA recognizes the likelihood of a relationship between the total load of SO_2 and NO_x and the acidity of precipitation. Therefore, TVA views acid deposition as a local, regional, national, and international problem that is not adequately addressed by the present requirements of the Clean Air Act.

If permitted to use the most cost-effective approach, TVA believes that its SO_2 emissions can be further controlled at a reasonable cost. In carrying out this least-cost approach, TVA believes that:

- Conservation should be recognized as a least-cost method of achieving compliance with any mandated reduction.
- Required emission reductions for the 31 acid rain control States should be proportional to each State's share of total SO_2 emissions from all sources for the 1980 base year.
- States should have the flexibility to allocate their emission reduction to any or all sources. Emissions trading and offsets for utility systems and other sources both within and between States should be allowed by the States. In determining a State's compliance in achieving its share of emission reductions, credit should be given to

reflect trading between sources in different States. NO_x and SO_2 emission reduction trading should be allowed on a two-for-one basis.

- After the States determine the SO_2 emission reductions that are required from each source, multiple sources such as utility systems should be free to choose the most cost-effective approach to obtain their allocated reductions regardless of State lines.

The availability and abundance of coal make it an important energy option. The environmental effects of increased coal combustion, however, stand as a principal deterrent to its use. TVA believes that the key to increased reliance on coal lies in minimizing those environmental effects by continued improvements in both control and combustion technologies. Accordingly, the Nation should maintain a high priority for research and demonstration of innovative technologies for the reduction of emissions from coal including atmospheric fluidized bed combustion.

ATTACHMENT 1

ASSUMPTIONS USED IN DEVELOPING COSTS OF ACID RAIN CONTROL BASED ON TOTAL SO_2 EMISSIONS

1. Utilities will be required to take all the SO_2 reductions for the State.

2. Each utility's share of the SO_2 reductions would be proportioned based on the total utility SO_2 emissions in the State.

3. TVA-required reductions in Tennessee, Alabama, and Kentucky can be taken anywhere on the system regardless of State lines.

4. Compliance will begin in 1995.

5. Each power plant has an economic writeoff period of 50 years. Limestone scrubber retrofits will be written off over the remaining economic writeoff period of each plant. A discount rate of 15 percent is used.

6. Limestone scrubber retrofits are evaluated at a capital cost of $200/kW in 1983 dollars, including interest during construction. A SO_2 removal efficiency of 85 percent is used. No major changes in scrubber costs are considered which could result from a major increase in demand.

7. The assumed price of low-sulfur coal does not include potential additional premiums due to a major increase in demand.

8. Compliance is met by a combination of least emissions dispatching, fuel switching, and limestone scrubbing.

9. No offsets or costs for additional controls for new sources are included.

10. Nuclear generation for each year will not drop below a level corresponding to an overall nuclear capacity factor of 60 percent. Normal hydro generation is assumed for each year.

11. No major unplanned outages at coal-fired plants where limestone scrubbers or lower sulfur coal is used are considered.

12. No transmission constraints which would prohibit or limit least emissions dispatching are included.

13. The minimum impact case is based on low load growth with a high level of conservation (20.9 billion kWh savings in FY 2000).

14. The major impact case is based on no additional nuclear capacity beyond Hartsville unit A 2. The minimum impact case is based on no additional nuclear capacity beyond Bellefonte unit 2.

15. Minimum cost case includes the cost incurred to protect against actual loads being higher than projected minimum.

16. Plants with limestone scrubbers for the maximum and minimum impacts are:

Reduction Goal	Maximum Impact	Minimum Impact
8 million tons	Cumberland (partial scrubbing)	---
10 million tons	Cumberland	---
12 million tons	Cumberland, Paradise unit 3, Gallatin (partial scrubbing)	Cumberland (partial scrubbing)

Cost to TVA of Various SO_2 Reductions if Allocation is Based on Total SO_2 Emissions in Each State[a,b]

National Reduction	Maximum Impact[c]			Minimum Impact[c]		
	Reduction in SO_2 Emissions from present stds	Capital Investment	Annual Cost	Reduction in SO_2 Emissions from present stds	Capital Investment	Annual Cost
	(1,000's tons)	($millions)	($millions)	(1,000's tons)	($millions)	($millio:
8 million tons	371	100	150	11	0	0
10 million tons	535	520	240	175	0	90
12 million tons	699	875	340	339	300	160

a. All costs in 1983 dollars.

b. Potential Rate Impacts on TVA System - TVA has the potential to achieve significant SO_2 reductions at relatively low cost by replacing coal-fired generation with generation from four nuclear units presently under construction. With projected load forecasts and the nuclear and coal-fired generation available to meet these projected needs, the expected rate impacts in 1995 would be as follows:

8 million ton reduction	0 to 2.6%
10 million ton reduction	2.5 to 4.2%
12 million ton reduction	4.5 to 6.0%

c. TVA's 1980 SO_2 emissions totaled 1.586 million tons compared to a peak emission level of 2.381 million tons in 1976. Under the maximum impact (high load growth scenario) SO_2 emissions would total 1.30 million tons in 1995 with present emission standards. Under the minimum impact (loads remain essentially unchanged from their present levels) in 1995, SO_2 emissions would total 0.94 million tons with present emissions standards.

8/9/83:E&PSS

Mr. FUQUA. Thank you very much, Dr. High.

And retroactively, without objection, permission to take photographs is granted.

Dr. Yeager?

STATEMENT OF DR. KURT E. YEAGER, VICE PRESIDENT AND DIRECTOR, COAL COMBUSTION SYSTEMS DIVISION, ELECTRIC POWER RESEARCH INSTITUTE

Dr. YEAGER. Thank you, Mr. Chairman, members of the subcommittees.

I'm Kurt Yeager, and I'm vice president and director of the Coal Combustion Systems Division of the Electric Power Research Institute.

The Institute was established by the electric utility industry in 1972 to conduct a broad research and development program for the entire electric utility industry and its ratepayers. As a part of this effort and through our R&D programs, we have been contributing significantly to the enhanced understanding of the acid rain issue. This commitment is based on the premise that our society will continue to have a strong environmental ethic and that the current environmental control capabilities must be improved by reducing both their cost and being able to better preserve the reliability of the powerplants that we have in place today.

For example, approximately 40 percent of the capital investment and 30 percent of the total cost of power for a new coal-fired powerplant is related to environmental control.

The urgent need for a technically sound basis to better evaluate the benefits and costs of reducing powerplant emissions suggests a comprehensive R&D program on all aspects of acidic deposition and, as such a national research program could provide the scientific and engineering data necessary to better guide the Nation in terms of the needs and appropriate timing of emission controls, it should be established.

In this regard, Electric Power Research Institute has suggested to both EPA and DOE an accelerated national acidic deposition research program, one that would be based on activities already underway at the Electric Power Research Institute and within the Federal agencies and one which has a high probability, in our judgment, of providing substantially better answers to the questions necessary to assess and deal with acid rain concerns in the next 5 years.

We estimate the total cost of such an accelerated R&D effort to be about $900 million over this 5-year time period. As a part of this, the ongoing EPRI commitment in that period to that issue will be $175 million. In addition, we will be spending an additional $300 million on science and technology related to coal-fired powerplant environmental issues other than acid deposition and advanced coal plant technology. The latter effort will include the fluid bed demonstration with TVA that Dr. High has mentioned, as well as a demonstration at a 100-megawatt scale of a gasification combined cycle powerplant scheduled to start operation at the Southern California Edison utility next year.

The overall program includes an accelerated effort to bring to fruition and better understand the issues of coal cleaning, furnace limestone injection, wet and dry scrubbing, water and solid waste disposal, and fluid bed combustion. These elements alone will be—consume $96 million from EPRI over the next 5 years.

The issue of acidic deposition has forced a national reexamination of control strategies aimed at reducing sulfur dioxide and nitrogen oxides. The objectives of more rapidly reducing the atmospheric loading of these pollutants has shifted the focus from new sources to the even more difficult problem of existing sources. The fundamental question, however, remains: How best to control the effects of intense urbanization in all its forms on the environment?

The key issue is not whether the various environmental impacts exist, but is there a control strategy which is technically capable of significantly reducing these phenomena and at a cost which is commensurate with the value of the benefits achieved.

A major consideration in any proposed action is the fundamental importance of coal to our Nation. More specifically, in terms of the eastern half of the Nation, which is a focus for most of the legislation in the acid deposition area, in 1980, 425 million tons of coal were consumed in 340 powerplants with a total capacity of 203,000 megawatts. That coal produced 80 percent of all the electricity generated in the eastern half of the Nation.

I'd like now to turn to some of the key issues, as we see them, in terms of the various options that exist for control and application to existing powerplants. I'd like to first start with switching to low-sulfur coal. Though not strictly a technical option, this control method is certainly an important and potentially cost-effective approach to dealing with some of the acid deposition concerns, and it has, however, associated with it a variety of technical and operational implications for the affected powerplants. These implications are often poorly understood and difficult to predict.

In addition, the limited public data base on the quality and availability of the Nation's massive coal resource limits accurate forecasts of the quantity, variability, and cost of coal, particularly of a low-sulfur content.

Two major coal supply regions stand to have a major reduction in their market due to low-sulfur coal switching, the Illinois basin and northern Appalachia, especially the two States of Ohio and Illinois. These, of course, create a number of political and institutional considerations which may limit the degree to which coal switching can occur, but, over and above those, there are also on the user side constraints. These include:

Particulate control efficiency. The switching to low-sulfur coal will typically affect the control capability of the existing electrostatic precipitators.

Combustion and handling characteristics moving from high-sulfur to low-sulfur coal, particularly western low-sulfur coal. There are some very distinct restrictions in terms of its acceptability in a powerplant for which it was not designed.

There is a good deal of concern over not only the increased cost of transport of coal, remembering that about 45,000 megawatts of the existing capacity in that eastern half of the Nation is classified as mine mouth, and, hence, it is using coal which is immediately

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available to it. Any switch from that would, of course, entail major transportation cost changes and the uncertainty of coal transportation costs today also creates uncertainty for the planners in the industry.

Therefore, the degree of coal switching will vary considerably based on the legislative requirements and the incentive provided. In our judgment, the possibilities of coal switching are probably, at least today, considerably less than often estimated by others.

Turning to coal switching—oh, excuse me—coal cleaning, this is perhaps, in terms of the economic order of dispatch of technology options, the one that would be turned to first.

Today essentially all commercial cleaning relies on physical separation methods. These methods are applied in varying degrees to over 50 percent of the high-sulfur steam coal used by the utility industry. This application has been primarily for economic reasons. Physical coal cleaning is most effective from a sulfur-removal standpoint for those coals that have pyritic sulfur content, particularly those again that are mined in northern Appalachia and the Illinois basin, where 25 to 35 percent of the total sulfur content may be removable.

However, much of this SO_2 removal potential has already been realized by the cleaning that is being done today. For example, over 50 percent of the sulfur-removal potential is already being achieved in the Illinois basin and about 30 percent in north Appalachia.

To achieve its full potential for this technology, the Electric Power Research Institute and the utility industry are active in the development of improved technology. In our case, this focuses on an Electric Power Research Institute commitment of more than $6 million a year plus significant cofunding by participating utilities. A focal point for this program is our coal-cleaning test facility located near Homer City in western Pennslyvania. Built entirely with private funds at a cost of more than $14 million, this is the newest and most advanced facility of its type.

Federal support recently in this area has been unfortunately small, and this is unfortunate I believe because the opportunities for technical progress in the area of steam coal cleaning are certainly substantial.

Some of the objectives that I believe should receive priority in this area are:

First, improved sampling and analytical procedures to insure confidence in our reserve quantity and quality estimates.

Techniques for improving the quality and making them more adaptable to eastern plants with respect to our western coal.

More efficient and economical fine coal cleaning.

As Dr. High mentioned, more economical chemical coal-cleaning techniques for more thorough sulfur and ash removal.

All of these provide a myriad of opportunities for improvements in the area of coal cleaning.

Unfortunately, in our judgment, under the limitations of most of the proposed acid deposition legislation, it is unlikely that the full potential of coal cleaning will be realized, because of its inherently low levels of sulfur removal, but if fully applied substantially over

the country, would be very large. To a large degree, this will not be permitted.

Flue gas desulfurization is the next option and certainly has to be the focus of most of the control approaches today. Because of its high SO_2 control efficiency, its emphasis for acid deposition control rapidly increases as control requirements become more stringent. Unfortunately, it is also one of our most expensive control options.

As of March 1983, there were 107 flue gas desulfurization units in operation on coal-fired powerplants in this country, with a total scrubbed capacity of 38,500 megawatts. An additional 64 units, representing 37,000 megawatts, were under construction or under contract. This represents a flue gas desulfurization commitment larger than the whole rest of the world combined. By 1992, this commitment will grow to include 32 percent of all coal-fired capacity, independent of any further acidic deposition control requirements.

Unfortunately, over the past decade, flue gas desulfurization systems to control emissions from coal-fired plants have proved to be among the most costly and least reliable pieces of equipment in the utility industry. As a result, the National Research Council in its 1980 report on this technology recommended that highest priority be given to improving the reliability of flue gas desulfurization or scrubbers for application to high-sulfur coal.

The EPRI R&D program to improve this unacceptable performance is the largest of its type in the country with $50 million invested to date and an equivalent amount planned for the next 5 years.

In this regard from the Federal standpoint, there was considerable applause when the Federal leadership for the development and evaluation of flue gas cleanup systems was transferred from the EPA to the Department of Energy, and this was applauded because it was felt this would promote resolution of many of the reliability, performance, and byproduct issues that limit the effectiveness of this mandated control technology. Unfortunately, to date, very few substantive programs have been implemented, although many opportunities exist and cosponsored projects with the utility industry and others have been developed for Federal participation.

Areas where concentrated effort is required, in our judgment, include:

First, with respect to the acid deposition issue, a substantially improved database for retrofit scrubber application.

Second, opportunities to improve the reliability of our existing lime and limestone scrubbing systems.

Opportunities to reduce the quantity and improve the quality of the byproducts associated with these systems.

It's important to note, for example, that one single scrubber on a typical 1,000-megawatt powerplant, burning 3 percent sulfur coal, can produce 1 square mile of waste byproduct 1 foot deep for every year of its operation. Added up over—up to 100,000 megawatts of retrofit scrubber capacity, demanded under much of the acid rain legislation, this adds up to a substantial land use penalty and an environmental side issue that should not be overlooked when judging these questions.

In terms of the retrofit issues of scrubbers, this is a much more difficult problem typically than associated with a new plant, be-

cause in a new plant one is able to design the scrubber and the powerplant as a system, an integrated system. In retrofit cases, we are forcing a complicated chemical engineering technology to be put on an existing plant that never was designed with that thought in mind.

These factors will typically increase the capital cost of a retrofit installation by 25 to 40 percent relative to a new plant. Costs well over $200 a kilowatt would be expected to be typical in a retrofit situation. Costs of over $300 a kilowatt have been documented.

The levelized costs, therefore, of these retrofit scrubber installations may typically rival the energy production cost, the busbar cost, of the electricity produced from these existing powerplants. In our judgment, therefore, scrubbers represent a bandaid solution, often creating as many problems today as they solve, and should be replaced by more effective technology as rapidly as possible, and we think that's where a lot of the R&D efforts should be going.

In the few minutes I have remaining I'd like to touch on two other options here that certainly are getting a lot of interest these days.

The first is furnace sorbent injection, otherwise known in some configurations as LIMB technology. This technology involves the injection of the dry sorbent, typically limestone, into the boiler for the absorption of the sulfur dioxide produced in the combustion process.

In the early 1960's this was initially tried as an effort to reduce SO_2 emissions without involving powerplants in complicated chemical flue gas treatment systems. The experience at that time was poor both from a performance as well as a reliability standpoint.

A variety of factors, however, contributed to renewed interest. These efforts, both in this country and in West Germany particularly, indicate the potential to achieve efficiencies for SO_2 removal on the order of 50 percent if the proper combustion and sorbent conditions can be properly controlled. The key to this seems to be in combining the technique with low-NO_x combustion, low-NO_x burners.

There are, however, a number of issues that should be resolved before commercial commitment to this technology can be confidently considered.

First, effects on furnace slagging and fouling, its effects on, in other words, ash composition, which degrade boiler availability and efficiency.

There's uncertainty in scaling up SO_2 removal capability, and currently the low-NO_x technology that seems to be associated with this would be applicable only to about a third of the pre-new source performance standard boilers that would be the target for this kind of an approach.

The large amount of limestone added will double or even triple the fly ash loading and, hence, affect the control technology, the particulate control systems on the back of the plant and also, accordingly, increase the ash disposal constraints.

Nevertheless, this has a number of potential merits to it, and as that result, the projects that we have underway are providing the groundwork for a major expansion by the Electric Power Research Institute and the utility industry. These will include participation

in the development, testing, and prototype field evaluations over the next 3 to 5 years. If that is successful, then we would propose to move on to large-scale demonstration later in this decade.

Two side issues that affect control technology that shouldn't be forgotten, one of which I mentioned earlier, and that is that little attention in this issue has been given to date to the secondary pollution questions of mitigation; that is, compliance with the various Clean Water Act and Resource Conservation and Recovery Act and the problems associated therewith as a result of the very large increase in both solid and liquid waste products created by these retrofit controls.

Second is the impact on particulate control on the electrostatic precipitators. Research in both those areas should be a focus of further effort.

Dr. High has mentioned the aggressive programs by the utility industry in the fluid bed combustion area. These results that have been forthcoming from the TVA effort on the 20-megawatt engineering prototype at Shawnee provided the basis for major demonstration that we will be initiating with TVA and Duke Power in the State of Kentucky.

In addition, we have four other projects that we have received proposals for, at least two of which we anticipate will proceed in the very near future, again based primarily on the private sector, almost totally on the private sector, initiative to move forward this technology.

One involves the retrofit of an existing boiler to a fluid bed to meet environmental controls, at a cost of less than $500 a kilowatt. Capital costs would be offset by resulting increase in generating capacity, fuel flexibility, and usefully extended plant lifetime, making it an even more cost-effective option in many situations than flue gas desulfurization.

Finally, in conclusion, I would like to say that if the Nation chooses to deal with the problem of acid deposition with—by requiring an arbitrary and rapid, very large, perhaps 10 to 12 million tons per year reduction in emissions, then it will be closing out a good number of these very valuable and important new technologies that are coming forth. In addition, a cost of $10 to $15 billion a year would be levied as a result of this kind of an approach, and the approach would restrict much of the more cost-effective options.

The potential for new types of generation, however, is great enough to introduce another tradeoff consideration. Although these technologies, such as fluid bed combustion, are several years from full commercial readiness, they allow emission reductions to follow naturally from the current economic and environmental evolution of coal-fired plants and would permit the Nation to apply the most cost-effective approach to emission reduction, and one which is aggressively being followed by the industry itself.

The premature use of what we would consider bandaid approaches would serve to freeze this technology in existing plants and would substantially continue the perceived conflict between coal and the environment.

In short, there is no shortage of opportunities to improve the efficiency, reliability, and environmental performance of coal combus-

tion. The successful achievement of the objectives laid out in this testimony, however, requires a much more constructive and coordinated partnership between Government and the private sector. This encompasses the planning and implementation of R&D, as well as the commercial transfer of results.

Speaking for the Electric Power Research Institute, we are prepared to do our part in continuing to formulate and implement these efforts.

I thank you very much.

[The prepared statement of Dr. Yeager follows:]

Testimony

By

Kurt E. Yeager, Vice President
Coal Combustion Systems Division
Electric Power Research Institute
Palo Alto, California

INTRODUCTORY STATEMENT

Mr. Chairman, Members of the Subcommittees:

I am Kurt E. Yeager, Vice President and Director of the Coal Combustion Systems Division of the Electric Power Research Institute (EPRI). I appreciate this invitation to meet with you to discuss the status of stationary source control technology research and development, particularly as it relates to the issue of acidic deposition.

The Electric Power Research Institute was established by the Electric Utility Industry in 1972 to conduct a broad research and development program for the entire electric utility industry and its rate payers. EPRI's R&D mission is to develop new and improved technologies for electric power production, delivery and use, and to help accelerate their availability to insure an adequate supply of economic, reliable and environmentally acceptable electricity.

EPRI is supported by voluntary contributions from investor-owned, government-owned and cooperatively-owned electric utilities across the nation. Through its R&D programs EPRI is contributing significantly to an enhanced understanding of the acid rain issue. This comprehensive R&D effort is carried out in close coordination with Federal programs and efforts by individual utilities.

This commitment is based on the premise that our society will continue to have a strong environmental ethic. Sound scientific data derived from independent, peer-reviewed research must be developed in a timely manner on topics of environmental concern. Current environmental control capabilities must be improved by reducing capital and operating costs and preserving or improving overall plant availability. Approximately 40% of the capital investment and 30% of the total cost of power for new, coal-fired power plants are related to environmental control. This includes sulfur dioxide (SO_2) scrubbing, particulate control, solid waste disposal, water treatment and plant cooling. These controls in their current form have a major impact on plant efficiency and have been one of the growing contributors to the loss of availability of coal-fired plants. Closely associated with this issue is the increasing difficulty in siting new utility and other energy facilities.

In addition, more information is needed to apply the potentially most cost-effective approaches to reduce emissions from power plants. Nevertheless, critical discussions are underway on the need for reducing emissions, especially from coal-burning power plants in the East and Midwest. Currently, proposed regulatory and legislative actions to control the emissions of power plants in this region could affect some 200,000 Megawatts of generation.

The urgent need for a technically sound basis to evaluate the benefits and costs of reducing power plant emissions suggests a comprehensive R&D program on all aspects of acidic deposition. A national research program could provide scientific data to guide

future decision makers on the needs and appropriate timing of emissions controls that should be established. Likewise, information on potentially attractive alternatives to current flue gas desulfurization practices could allow the selection of considerably more cost-effective options.

Since its inception, EPRI has worked with the Department of Energy (DOE and its predecessor agencies), the Environmental Protection Agency, (EPA), and other Federal agencies with energy research and development responsibilities to achieve mutual technical objectives. In the spirit of that long-standing cooperative relationship, EPRI has suggested to EPA and DOE an accelerated national acidic deposition program. This builds on activities already underway at EPRI and within the Federal agencies, and has a high probability of providing better answers to the questions necessary to assess and deal with acid rain concerns in the next five years.

The proposed program does not rely on a "demonstration" project to reduce emissions during the period because such an effort would be costly and require at least 10 years (5 years to reply and at least 5 years of monitoring) to provide statistically significant results, if done at a meaningful scale.

We believe the proposed R&D effort will yield equivalent results at a fraction of the cost and provide the information EPA needs to take appropriate action to protect or restore any resources determined to be a risk from urban, industrial or utility emissions.

We estimate the total cost of the accelerated R&D effort to be about $900 million over the five-year period. In addition to the EPRI commitment, we expect utilities to contribute significantly to the emissions control work and government, federal and state, to the ecological effects, atmospheric science and resource management components. We will spend an additional $300 million on science and technology related to coal-fired power plant environmental issues and advanced coal plant technology during this same period. This latter effort includes a 160 MW demonstration of fluidized bed combustion (FBC) with TVA for startup in 1989, and a 100 MW demonstration of a gasification-combined cycle (GCC) power system scheduled to start operation at Southern California Edison next year.

We believe that if the utilities and the government collaborated on follow-on efforts to these pioneer plants, the technology could be commercially available for plants ordered in the mod-to-late-1990s. The environmental benefits accruing to the nation's water and land, as well as its air, would far surpass those to be achieved by massive expenditures on older coal-fired plants.

Timing of some of the key milestones in the environmental control portion of this accelerated national program is shown in Figure 1. Funding requirements for this effort are illustrated in Table 1 and total $480 million over the 1984-1988 period. This includes $96 million planned by EPRI.

BACKGROUND

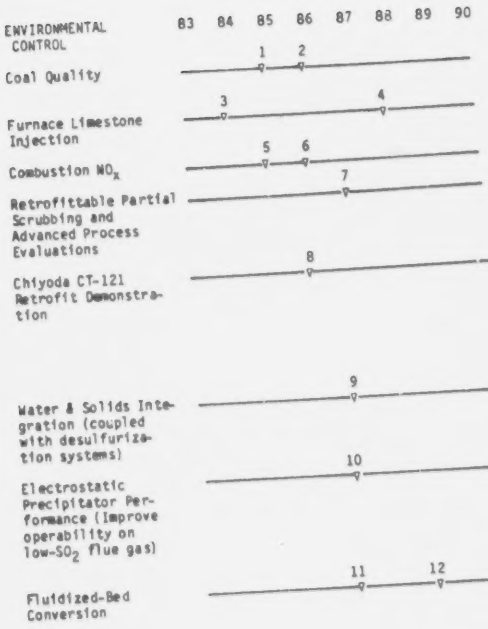
The issue of acidic deposition has forced a national reexamination of control strategies aimed at reducing sulfur dioxide (SO_2) and, to a lesser extent, nitrogen oxides (NO_x). The objective of more rapidly reducing the atmospheric loading of these pollutants has shifted the focus from new sources to the even more difficult problem of existing sources. The fundamental question, however, remains how best to control the effects of intense urbanization on the environment. Scientific uncertainty surrounding many aspects of these effects complicates the decision of whether or when to require further emission controls and from what sources. Unfortunately, political pressure to force rapid judgements on one or another emitting sector without the means for accurate, objective consideration of the total cause and effect is unlikely to be effective. Substantively, however, these judgements are sure to impose major costs on the affected sectors and our society as a whole. The key question is not whether the environmental impacts exist, but whether there is a control strategy which is technically capable of significantly reducing these phenomena at a cost commensurate with the value of the benefits to be achieved.

In response to the current politically charged issue of acidic deposition, proposed legislative action has targeted on coal-fired plants because of their role in SO_2 emissions. Collectively they emit a little over one-half of the man-produced SO_2 and one-fourth of the man-produced NO_x.

A major consideration in any proposed action is the fundamental importance of coal to our nation. For example in 1980, the baseline year for much of the proposed acidic deposition legislation, coal fueled 60% of all electricity produced in the United States. More specifically, in the 31 state eastern half of the nation which is widely being considered for more stringent emission control measures to combat acidic deposition, 425 million tons of coal were consumed in 340 power plants with a total capacity of 203,000 MW. This coal had a reported average sulfur content equivalent to 3.2 pounds of SO_2 per MMBtu and produced 80% of all electricity generated in the eastern half of the nation.

During the period of 1975-1980, SO_2 emissions in this 31 state region were already reduced by 2.4 million tons per year (MMTPY). This included 20% to 40% reductions by utilities in Michigan, Kentucky, Ohio, Illinois, Tennessee and Alabama, primarily through reductions in the sulfur content of the coal consumed.

Whether or not widespread additional action is taken against existing coal-fired power plants, no single solution will satisfy the widely varying conditions in these plants. There are several basic control approaches and any number of technical variants under



Representative Milestones

1. Complete characterization of 25-30 Bituminous Coals
2. Complete testing and guidelines for beneficiation and blending of low rank coals.
3. Complete pilot-scale development.
4. 20-100 MW prototype results complete. Early results from multiple full-scale demonstrations.
5. Complete test of prototype wall-fired burners.
6. Initial results from multiple full-scale tangential-fired and wall-fired burner demonstrations.
7. Results from in-duct spray drying and other partial SO_2 removal demonstrations.
8. Operating data available from 150-300 MW Demonstration.
9. Initial results from full-scale integrated system demonstrations.
10. Results available from full scale field evaluations.
11. Startup of retrofitted FBC boiler.
12. Complete performance testing of FBC Retrofit boiler. Design and operational data available.

MILESTONES ACCELERATED CONTROL TECHNOLOGY PROGRAM

FIGURE 1

development or in use today. Possible options for SO_2 reduction include:

- o Fuel changes - coal cleaning, fuel switching and blending, coal conversion.
- o Process changes - flue gas desulfurization, fluidized bed combustion and furnace sorbent injection.
- o Operational changes - emission trading/offsets, retirement of units, emission dispatch, derating and load reduction.

First they differ significantly in their control potential. Second, their costs do not neatly match the progression of control efficiency. Third, is the variety of functional criteria that determine technical and economic applicability: coal quality, plant design, furnace geometry, component capacities, duct configuration, space requirements, land and water availability, etc. This third category is a particularly important constraint when considering the retrofit of control technology to existing plants. Unlike new plants where power plant and emission controls are designed as an integrated system, retrofit requires adaptation of emission controls to a plant typically not designed for such modification. This can lead to compromise and additional expense as discussed in the following sections of this testimony.

Over the past decade, EPRI, with the utility industry and its suppliers, has aggressively pursued the development of emission control technology with improved reliability and cost to meet the requirements on electric power generation. It is from this foundation that EPRI is able to address the issues surrounding accelerated retrofit requirements and develop the perspective reflected here.

SWITCHING TO LOW-SULFUR COAL

Electric utilities are the largest consumers of coal in the United States, and are projected to account for over 70% of the market in 1995. Any major shift in demand from this sector, therefore, will have a heavy impact on the coal industry. Because of the inherently higher sulfur content of current coal production, only 95 million tons per year (MMTPY) or 17% of the coal consumed by the utility industry in 1980 had a sulfur content less than 1.2 lb per MMBtu. Most of this was consumed in the western half of the nation where 90% of the reserves of this very low sulfur coal are located.

Although not strictly a technical option, this control method has a variety of technical and operational implications for the affected power plants. These implications are often poorly understood and difficult to predict. In addition, the limited public data base on the quality and availability of the nation's massive coal resource limits accurate forecast of the quantity, variability and cost of coal, particularly of low-sulfur content.

Coal switching may be attractive because in many situations it is the most cost-effective strategy to reduce SO_2 emissions. However, the first consideration must be technical feasibility. When that answer is determined, the utility must then quantify the cost.

Table 1

TECHNICAL RESPONSES - SCHEDULE AND FUNDING REQUIREMENTS ($M)

Response	84	85	86	87	88	Total	Proposed 1984-1988 EPRI Plan
1. COAL QUALITY IMPROVEMENT							
Conventional Coal Cleaning	5	10	10	10		35	
Advanced Coal Cleaning	6	6	6	6	6	30	40.0
Low Rank Coal Beneficiation	2	3	3	4	3	15	
Coal Characterization	5	5	5	5	5	25	
2. FURNACE LIMESTONE INJECTION							
Prototype Tests (3)	6	8	6	2	1	23	
Full-Scale Demos. (2)	2	15	20	10	3	50	11.0
Pilot Studies	6	4	3	2	1	16	
3. COMBUSTION NO_x							
Full-Scale Demos. (3)	5	14	8	7	2	36	7.0
4. RETROFITTABLE PARTIAL SCRUBBING							
Prototype Tests	3	4	2	1		10	
Economic Tradeoffs	1	1	1	1	1	5	5.0
Full-Scale Demo	5	15	15	5	5	45	
5. CHIYODA CT-121 RETROFIT							
150 MW Demonstration	6	12	12	5		35	.5
6. ADVANCED PROCESS EVALUATIONS							
Pilot Evaluations	3	5	5	5	2	20	4.0
Engineering/Economic Eval.	1	1	1	1	1	5	
7. WATER & SOLIDS INTEGRATION							
Full-Scale Demos. (2)	5	10	15	10	5	45	8.0
8. ELECTROSTATIC PRECIPITATOR PERFORMANCE IMPROVEMENT							
Upgrade Technology	3	3	3			9	5.0
Field Evaluation		2	2	7	5	16	
9. FLUIDIZED BED CONVERSION							
100 MW Demonstration	20	30	5	3	2	60	15.0
Total	84	148	122	84	42	480	95.5

The principal cost elements of coal switching are any increase in delivered cost of coal plus the cost of modification and/or loss of generating capacity. The latter is particularly important when considering switching to, or blending, low-sulfur western coal.

There are, of course, major political and institutional considerations which may outweigh cost-effectiveness in the decisions to switch to low-sulfur coal. Many of the states most affected by proposed acid rain legislation are producers as well as consumers of high-sulfur coal and have a jobs-and-income interest in maintaining utilization of local coals. In addition, many utilities either operate captive mines or may have difficulty terminating long-term coal supply contracts for high-sulfur coal that generally contain "take-or-pay" provisions. Over 40,000 MW of capacity is also classified as mine-mouth which will substantially increase the cost penalty of coal switching.

It is also important to recognize that coal switching can be limited by very stringent control requirements. For example, it is unlikely that much switching could occur at SO_2 emission levels below 1.5 lb/MMBtu. This would require an eastern bituminous coal of less than 0.8% sulfur content. Eastern coal of this sulfur quality is extremely limited and used mainly for the demands of the steel industry. On the other hand, use of western low-sulfur coal would be ineffective in meeting these very low emission levels because of the blending and derating constraints on its use in most midwestern and eastern power plants.

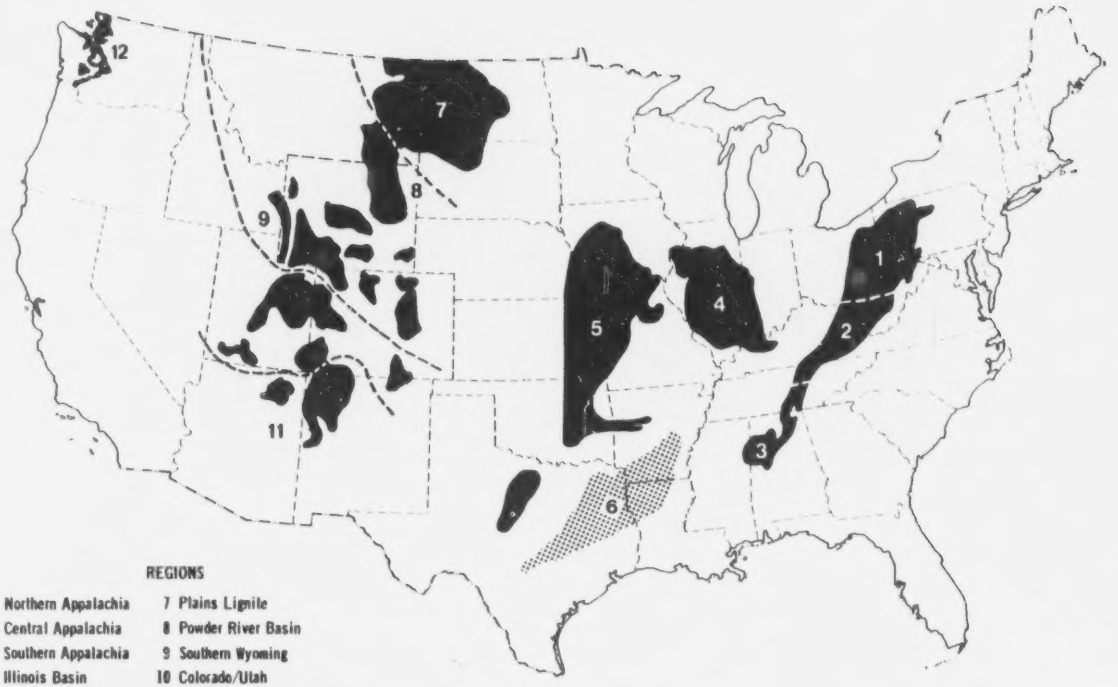
In the East, reserves of low-sulfur steam coal are found primarily in the central Appalachia region (Figure 2). This region includes southern West Virginia, eastern Kentucky, Virginia and northeastern Tennessee. By comparison, with the exception of the Western Interior region, all of the western coal regions produce low-sulfur coal. Much of this, however, is of lower heating value, subbituminous quality. Utilities which have already had to meet lower sulfur limits typically switched from northern Appalachia and Illinois Basin high-sulfur coal to Central Appalachia and the Powder River Basin. The higher heat content southern Wyoming and Colorado/Utah regions may also play a role in supplying utilities switching to low-sulfur coal as a result of any acid rain legislation.

The two major coal supply regions which stand to lose markets due to low-sulfur coal switching are the Illinois Basin and Northern Appalachia, especially the states of Illinois and Ohio. These regions each sell 100-120 million tons per year (MTPY) of coal to electric utilities today. It is important to note the effective regional restriction on coal distribution from these two regions. Over 85% of Illinois Basin coal and 95% of northern Appalachian coal is sold in the producing or adjacent states.

The low-sulfur coal markets in the western and eastern United States are very different. The West has large reserves of low-sulfur coal and these reserves can be recovered through lower cost surface

FIGURE 2

Coal Supply Regions



REGIONS

1 Northern Appalachia	7 Plains Lignite
2 Central Appalachia	8 Powder River Basin
3 Southern Appalachia	9 Southern Wyoming
4 Illinois Basin	10 Colorado/Utah
5 Western Interior	11 Arizona/New Mexico
6 Gulf Lignite	12 Washington

mines. In the East, low-sulfur coals are relatively scarce, much of the original reserve base has been depleted and low-sulfur reserves are in seams which only higher cost, labor intensive, underground mines can recover. As a result, low-sulfur western coal prices would probably remain fairly stable, in real terms, in the face of increased demand resulting from any acid rain legislation. By comparison, there already is a distinct premium for eastern low-sulfur coal, based on export and steel making demands as well as competing utilities. This price premium would be likely to increase further in the face of increased demand.

Because of over-forecasting of coal demand in the late 1970's the coal industry appears able, through existing investments, to meet the low-sulfur coal demand that may occur as a result of acid rain legislation. In their 1980 Annual Report to Congress, for example, DOE estimated 1990 eastern coal production to be 804 million tons, with western production at 608 million. In the 1982 version, DOE reduced its 1990 eastern forecast to 692 million tons, with western production at only 388 million.

Several other factors do, however, constrain the ability of utilities to practically swtich to low-sulfur coal. These include:

1. Particulate Control Efficiency. Many older power plants are already only marginally in compliance with particulate limitations. As these limits have tightened over the years, these plants are likely to require substantial modifications to achieve compliance if switched to low-sulfur coals.

2. Combustion and Handling Characteristics. Pulverized coal boiler design heavily depends on the characteristics of the coal to be burned. Thus, the design will generally limit the boilers' capability to burn low-sulfur western coals unless it is modified at high capital cost or derated. Moisture, volatile matter, ash composition and heat content are key characteristics affecting combustion. Collectively these issues will constrain the extent to which low-sulfur western coal can be used in existing midwestern and eastern power plants.

   The issue of coal grindability is also serious for older boilers designed to burn relatively soft northern Appalachia coal. Low-sulfur coal from central Appalachia, or the West, is usually much harder. This will force a utility converting to this coal to add pulverizer capacity or derate the boilers. Subbituminous Western coals also tend to produce much higher levels of dust in unloading, reclaiming and transferring. Hence, handling system modifications are required to prevent explosions and fires.

3. Transportation. Utilities which are burning high-sulfur coal today are burning it because it is the most economic coal available on a delivered cost basis. For power plants located in or near the high-sulfur coal fields of the Illinois Basin and Northern Appalachia, this coal has significantly lower transportation costs than coal from other supply regions. A major

part of the economics of choosing coal switching or blending will be the increased coal transportation costs and the potential for further escalation.

The following table provides two examples to put these cost factors affecting coal switching and blending into perspective. These are intended to be illustrative only of the situation which might face typical plants using high-sulfur Illinois Basin or north Appalachia coal.

	Illinois Basin Power Plant, Blending Powder River Coal*	N. Appalachia Power Plant Switch To Central Appalachia Coal
	(1983 $)	
Capital Cost ($/kW)		
Boiler	20	0
Auxiliaries	20	5
ESP	40	30
Derating	50	0
	130	35
($/MMBtu)		
Annualized Capital	0.45	0.12
Incremental Fuel Cost	0.28	0.90
Operating Cost	0.03	0.02
Total Cost	0.76	1.04
Resulting SO_2 Emissions Rate (lb/MBtu)	2.4	1.5
Cost/Ton SO_2 Removed ($)	657	595

* 60% Powder River Coal

The extent of coal switching will, therefore, vary considerably based on the legislative requirements and incentives provided. EPA has, for example, concluded that acid rain legislation could produce a 60 MMTPY increase in central Appalachia coal production and a 100 MMTPY increase in western coal production with corresponding production displacement (adjusted for coal heat content differences) primarily in the Illinois Basin and north Appalachia coal regions. Given the technical, institutional, and economic constraints on coal switching, it is unlikely that this degree of shift will actually occur. Coal switching is more likely to result in a net increased low-sulfur coal production increment on the order of 50 to 100 MMTPY. This could be expected to result in a reduction in annual SO_2 emissions of 1.5 to 3 MTPY.

COAL CLEANING

Coal is a complex heterogeneous substance containing significant quantities of inorganic impurities including sulfur. The relative amounts of these impurities, the manner in which they are included in the coal structure, and the degree to which they can be removed vary widely among different coals.

The sulfur content of U.S. coals ranges from less than 1 to more than 7 percent. It appears in two principal forms: organic sulfur and mineral sulfur in the form of pyrite. Organic sulfur, comprising 30 to 70 percent of the total sulfur content of most U.S. coals, is an integral part of the coal matrix and can be removed by chemical modification of the coal structure. Pyritic sulfur occurs in coal as discrete mineral particles, often of microscopic size. Pyrite is a heavy mineral with a specific gravity of 5.0; coal has a maximum specific gravity of only 1.7. To take advantage of this difference, the most wide using cleaning techniques for coal utilize density separation methds at specific gravities of 1.4 to 1.6. The degree of desulfurization obtained by crushing and specific gravity separation depends on the initial sulfur level in the raw coal, the ratio of pyritic-to-organic sulfur, the size distribution of pyrites, and the cleaning techniques used. In general, pyrite removal increases with increased crushing and lower specific gravity of separation.

Commercially, coal today is physically cleaned by crushing the mined coal to a point where some of the mineral impurities are released from the coal structure. The minerals (including pyritic sulfur) and coal particles can then be separated by techniques that rely on differences in the specific gravity, surface properties or magnetic properties of the particles. The process is made up of several unit operations tailored to the characteristics of the coal and its end use. These encompass three primary steps: crushing, separation and dewatering/drying.

While several common physical coal cleaning mechanisms are used, there is no universal approach to coal cleaning plant design. This is a result of the substantial variability in the physical and chemical composition of coal between and within seams. Therefore, a given preparation process that is effective with a coal from one seam may be inappropriate with a coal from another seam to achieve a comparable level of cleaning. As particle size decreases, the chances for separating the coal and impurities increase, but processing costs increase as well. For this reason and to avoid losses during transportation, the cleaning process for most seam coal involves crushing to a top size of 1.5 inches. Coal cleaning or beneficiation, can be separated by levels; typically (a) no preparation or breaking, (b) partial cleaning, and (c) intensive cleaning. To achieve maximum practical pyritic sulfur removal, this testimony considers only coal cleaning plants built or upgraded to an intensive cleaning capability.

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In chemical cleaning, more complete removal of ash and organic, as well as pyritic sulfur from the coal, is possible. Chemical techniques range from leaching of inorganic matter to solvent refining which dissolves the coal under increased temperature and pressure. The much higher cost of these generally developmental chemical cleaning processes has, unfortunately, largely discouraged their commerical consideration for use on utility steam coal.

Today, essentially all commercial coal cleaning relies on physical separation methods. These methods are applied in varying degrees today to over 50% of the high sulfur steam coal used by the utility industry. This application has been primarily for economic reasons. First to reduce transportation and waste disposal costs and second to reduce operating and maintenance costs in the boiler and coal handling equipment caused by uncombustible impurities. The increasing variability in coal quality and the resulting impact in power plant performance and availability has also led the utility industry to increase its attention to quality control by increased cleaning of its coal feedstock. The third major benefit of coal cleaning, and the object of this testimony, is emission reduction. Physical coal cleaning is most effective for bituminous coal with high pyritic sulfur content, particularly those mined in north Appalachia and the Illinois Basin where 25% to 35% of the total sulfur content can typically be removed. However, much of this SO_2 emission reduction potential has already been realized in the cleaning done today. For example, over 50% of the sulfur removal potental is already being achieved in the Illinois Basin and about 30% in north Appalachia. On the other hand, physical coal cleaning to remove sulfur is typically ineffective for the low-sulfur southern Appalachian and western coals. As a result, cost effectiveness varies from about $200 per ton of sulfur removed for some Illinois Basin coals (Illinois, Indiana, west Kentucky) to over $2000 per ton of sulfur removed from some low-sulfur coals.

The opportunities to improve sulfur and ash removal through coal cleaning, therefore, vary widely and depend on upgrading existing coal cleaning facilities as well as installation of new plants. The limiting cost factor in physical coal cleaning is the loss of coal during the cleaning process and the effective heating loss from increased moisture content. To achieve its full potential, EPRI and the utility industry are active in the development of improved coal cleaning technology plus characterization of steam coal cleanability and cost. The emphasis is on finer (smaller particle size) coal cleaning and drying techniques to improve coal recovery and thus reduce cost. It is also anticipated that finer cleaning will require even greater R&D emphasis as we try to achieve more ash and pyrite liberation.

This coal cleaning R&D program invoives an EPRI commitment of more than $6 million a year plus significant cofunding by individual utilities, and others. A focal point for this program is the EPRI Coal Cleaning Test Facility(CCTF) located near Homer City, Pennsylvania, and operating since 1981. Built entirely with private funds at a cost of $14 million, this is the newest and most advanced facility of its type. Based on commercial equipment, the CCTF is demonstration scale with a maximum coal handling rate of 20 tons

per hour. Unlike other test facilities, it is not dedicated to a single cleaning process but can accommodate a variety of projects resulting from EPRI research as well as others in the coal R&D community. Other objectives include operation training and development of a coal cleaning data base for the utility industry.

Cleaning operations currently being evaluated at the facility include heavy media cycloning, water only cycloning, tabling, and froth flotation. Fine coal and refuse dewatering can be accomplished by either centrifugation or filtration. Its flexibility permits simulation of more than 50 commercial cleaning plant system designs. The result: an appraisal of not only how well each unit operation performs individually but also how well it integrates with the rest of the plant.

DOE support for this area has been small in recent years. This is particularly unfortunate because the opportunities for technical progress is very large relative to the investment required. In addition, such progress can be rapidly translated into substantial improvements in both the quality and quantity of coal utilization. DOE is also blessed with very capable laboratories and staff to perform these efforts.

Priority objectives for research and development to improve coal cleanability forecasts and technology include:

- Improved sampling and analytical procedures to insure confidence in our reserve quantity and quality estimates.
- Techniques for cleaning lower rank Western coals.
- More efficient and economical fine coal cleaning and recovery techniques.
- More economical chemical cleaning techniques providing thorough sulfur and ash removal.
- Automated cleaning process control and on-line continuous measurement techniques.

There is also substantial uncertainty concerning the performance and utilization of existing coal cleaning facilities. As a result, accurate determination of the opportunities to use excess capacity in existing cleaning plants and upgrade their sulfur removal capacity is difficult. Because of the heterogenous nature of coal, the cleanability and associated cost and benefits can vary widely. For the high-sulfur north Appalachian and Illinois Basin coal producing regions typical costs are indicated below:

	New Intensive Cleaning Plant*	Upgrade Plant To Intensive Cleaning Capability*
Capital cost ($/kW)	45	20
Levelized Cost ($/MBtu)	0.3-0.45	0.14 - 0.17
Cost per ton clean coal	($) 7 - 10	3 - 3
Cost per ton SO_2 removed	($) 400 - 800	500 - 800

* 1000 tons/hr capacity, 1983$, 30 year levelized cost basis capacity factor 40%, availability 80%.

In some cases, further savings in addition to transportation and waste disposal may also be achieved through improved boiler reliability and reduced maintenance. Much remains to be done, however, in correlating these effects on a site specific basis. On the other hand, it should also be noted that the large majority of coal cleaning is accomplished by the coal producer, at or near the mine. As a result, the actual price premium associated with cleaned coal may be substantially higher than the actual cost of cleaning, depending on the market factors existing at the time.

Unfortunately, under the limitations of most of the proposed acid deposition legislation, it is unlikely that this SO_2 reduction potential of coal cleaning will be realized. SO_2 reduction requirements of 50% or more and/or very low emission limits exceed the cleanability potential of coals currently burned in the affected region. Alternatively, the relatively small quantity of low-sulfur coal used has an inherently low sulfur removal potential. As a result, it is unlikely, unless more compliance flexibility is encouraged, that additional SO_2 reduction through expanded coal cleaning will exceed 0.5 MMTPY although the potential exists for 1.5 MMTPY or more of SO_2 reduction.

FLUE GAS DESULFURIZATION

Flue gas desulfurization (FGD), so-called wet scrubbing, has been defined by EPA as the Best Available Control Technology (BACT) for control of SO_2 emissions. Because of its high SO_2 control efficiency, its emphasis for acidic deposition control rapidly increases as control requirements become more stringent. Unfortunately, it is also one of the most expensive control options. More than 100 FGD processes have received consideration for use in removing SO_2 from flue gases, but only three or four are now commercially applied in the United States. All use aqueous scrubbing and generally require disposal of a waste product. As of March 1983, there were 106 FGD units in operation with a total scrubbed capacity of 38,500 MW. An additional 64 units are under construction or contract totaling 37,000 MW. This represents an FGD commitment greater than the rest of the world combined. The operational FGD capacity today controls 15% of all coal-fired generation and will grow to 32% by 1992, independent of any further acidic deposition control requirements.

The most widely used FGD approach is scrubbing with a slurry of lime or limestone and production of a throwaway byproduct. Over 80% of the installed and planned FGD capacity is of this type. Unfortunately, over the past decade FGD systems to control SO_2 emissions from coal-fired power plants have proved to be among the most costly and least reliable pieces of equipment in the utility industry. Accounting for as much as 25% of the capital and operating cost of a new power plant, these FGD systems on high-sulfur coal have typically experienced severe corrosion and plugging problems and have seriously detracted from the availability of the affected power plants. The $175/kW or more capital cost of FGD on a new plant is exceeded only by the cost of the boiler itself. Maintenance cost for the FGD system is two to twenty times the maintenance cost for the rest of the power plant. As a result, the National Research Council in its 1980 report on FGD technology recommended that highest priority be given to improving the reliability of FGD systems for application to high-sulfur coal. Although average FGD availability on medium and high-sulfur coal has increased from 53% in 1978 to 85% in 1981, this is still not consistent with utility requirements and represents a substantial loss in available electric generating capacity.

The EPRI R&D program to improve this unacceptable FGD performance is the largest of its type in the country with $50 million invested to date and an equivalent amount planned for the next five years. It emphasizes failure cause analysis, improvements in process chemistry, materials of construction, water and energy use, hardware design, plus performance data collection and interpretation. In addition, EPRI is actively pursuing the development of improved FGD designs capable of reducing byproduct production and substantially improving cost and reliability. These include demonstrations of both the Aqueous Carbonate Process (with EPA, ESEERCO & NYSERDA) and the Chiyoda 121 Process.

Several years ago, Federal leadership for the development and evaluation of flue gas cleanup systems was transferred from EPA to DOE. This was applauded as a progressive step promoting resolution of reliability, performance and byproduct issues limiting the effectiveness of mandated control technology, particularly FGD. This need is punctuated by the fact that 40 percent of the capital cost of a new coal-fired power plant is typically for compliance with environmental requirements. To date, however, very few substantive programs have been implemented, although many opportunities for flue gas cleanup technology improvement exist and cosponsored projects with utilities and others have been developed for DOE participation.

The operational limitations associated with the required flue gas cleanup technology, together with uncertainty over the future course of environmental regulatory requirements cause the greatest impediment to the increased use of coal. It is, therefore, disturbing that the government has maintained a position which effectively discourages cooperation with the private sector in the solution of technical issues created by the technology it has mandated.

Recommended areas where concentrated R&D effort is required include:

- Develop an improved design basis for retrofit scrubber application.
- Improve the reliability of existing slurry (lime/limestone) scrubbing-systems for sulfur dioxide removal.
- Reduce the quantity and improve the quality of disposable byproduct associated with slurry scrubbing systems.
- Demonstrate advanced regenerable flue gas desulfurization systems which minimize the use of expensive reagents, produce marketable byproducts and rely entirely on coal.
- Develop and demonstrate compact "urban scrubber" flue gas cleanup systems supporting the conversion of existing oil-fired power plants to coal.
- Develop simplified flue gas cleanup systems capable of simultaneous removal of sulfur and nitrogen oxides.
- Develop entirely dry, retrofitable FGD processes which can be integrated with existing particulate emission control systems.

The effectiveness of government funding in this category will depend on a much more aggressive R&D policy which focuses on the key problem areas indicated and commits jointly with the private sector to the major technology projects required for their resolution. By comparion, EPRI alone is investing annually in this area about five times the FY1983 DOE request.

Retrofitting an FGD system to an existing power plant as proposed under acidic deposition legislation, is typically more difficult and expensive than on a new power plant. This results from the necessity to apply a complex chemical process to a power plant not originally designed to accept it. This increased difficulty and cost is affected by a variety of site specific factors including:

- Remaining Plant Life
- Accessibility
- Demolition Requirements
- Undergrounds
- Particulate Control Efficiency
- Ductwork
- Fan Capacity
- Stack Condition
- Waste Disposal Siting

These factors will typically increase the capital cost of a retrofit FGD installation by 25% to 40% relative to a new plant. However, many instances have been identified where a 100% or more cost increase is expected. These estimates do not include the added cost of power plant downtime while ducts and stacks are being prepared for the FGD system.

The levelized cost of these retrofit FGD installations may typically rival the present busbar cost of the electricity produced by the power plant. This is especially influenced by the remaining life of these plants. Previous cost estimates of acidic deposition control strategies have typically underestimated these retrofit FGD costs.

These known problems with FGD systems make them an expensive and difficult option for retrofit to existing plants. In our judgement they represent a band-aid solution, often creating as many problems today as they solve and should be replaced by more effective technology as rapidly as possible. Adding them to older plants is generally uneconomic and should be avoided except in cases where the risk to the ecosystem is found to be unacceptably large and no other alternative is available. Current R&D efforts by EPRI and others may reduce these impacts but no opportunities are available to produce a radical improvement in the near future for FGD.

Typical retrofit planning values for both limestone scrubber and lime spray dryer FGD systems on a 500 MW power generating unit burning 2.5% sulfur content coal with a 60% capacity factor and 15 years remaining life are shown below.

	Limestone Scrubber (1982 $)	Lime Spray Dryer
Capital Cost	230 $/kW	190 $/kW*
- Total Levelized Cost	20 mills/kWh(1)	21 mills/kWh(1)
- Energy Use	3-5% of plant input	1-2% of plant input
- Land Use (fixed landfill)	240 acre-ft/yr(2)	250 acre-ft/yr(2)
- Solid Waste	220,000 TPY(2)	240,000 TPY(2)
- Water Use	500-2500 GPM	400-600 GPM
FGD Availability	80-90%	80 - 90%
- Unit Reliability Impact	-5%	-5%
- SO_2 Removal Efficiency	80 - 90%	60 - 85%
- Cost/Ton SO_2 Removal	1000 - 1150$(1)	1000 - 1300 $(1)

* Includes Fabric Filter Baghouse

(1) Does not include cost of any lost generating capacity.
(2) Wet disposal may double these quantities.

The cost of FGD is much more dependent on the volume of flue gas being scrubbed than on the SO_2 removal efficiency required. It is, therefore, more economical to scrub a portion of a unit's flue gas to high efficiency than all the flue gas at low efficiency. Physical coal cleaning may also be usefully applied in conjunction with FGD in many situations. The value is less in reducing the overall capital cost, however, than improving scrubber reliability and operating cost.

FURNACE SORBENT INJECTION

one of the more publicized techniques under development for reducing SO_2 emissions from existing boilers is Furnace Sorbent Injection, also known in one configuration as LIMB.

Furnace sorbent injection for SO_2 control consists of injecting a dry pulverized calcium-based material directly into the furnace cavity of a coal-fired boiler. Upon entering the furnace the sorbent material decomposes into lime (calcination) which reacts in suspension with SO_2 to form solid calcium sulfate. The calcium sulfate and unreacted sorbent particles are then removed along with the fly ash in the particulate control device.

The process is entirely dry and utilizes conventional equipment for the storage, handling and injection of the sorbent materials. Limestone is the most abundant sorbent material available throughout the United States and is the preferred dry sorbent material from the standpoint of availability and cost.

In the early 1960's, the concept of direct sorbent injection into the furnace of a utility boiler originated as a means of reducing SO_2 emissions without involving utility companies in complicated chemical flue gas treatment systems. The low and variable removal efficiencies combined with boiler operational concerns essentially discredited this approach and by the early 1970's, the development of wet SO_2 scrubber systems became the primary focus of SO_2 control efforts.

The relatively low SO_2 removals and poor sorbent utilization experienced in the early tests are now believed to be attributed to three factors:

(1) When the sorbent was injected into the burner flame zone, the high temperatures tended to deactivate the lime (i.e. "dead-burning").

(2) When the sorbent was injected away from the flame zone in the upper furnace region, inadequate mixing with the SO_2-bearing combustion products led to low sorbent utilization.

(3) Poor fundamental understanding of the SO_2 capture process prevented optimation of the process for the various site-specific boiler, burner, fuel and sorbent conditions encountered.

A variety of factors have contributed to the renewed interest in furnace sorbent injection. Foremost among these are the recent experimental data here and in West Germany, that indicate the potential to achieve SO_2 removal efficiencies on the order of 50% if combustion and sorbent conditions can be properly controlled. A calcium to sulfur ratio on the order of three is anticipated to achieve this level of control. Along with this understanding is the realization that new combustion systems being developed to control emissions of nitrogen oxides (NO_x) appear to help achieve these necessary conditions. In particular, the lower combustion temperatures and controlled mixing conditions associated with these

systems may provide new opportunities to optimize the sorbent injection process and also provide the added benefit of simultaneous NO_x and SO_2 control. A final factor contributing to the resurgence of interest in dry sorbent injection is the growing incentive for low-cost incremental SO_2 controls, applicable to existing power plants.

There are a number of issues that must be resolved before commercial commitment to furnace sorbent injection technology can be confidently considered. The major issues identified by both U.S. and West German developers regarding utility boiler retrofit applications are:

(1) Effects on furnace slagging and fouling which degrade boiler availability and efficiency.

(2) Uncertainty in scaling up laboratory SO_2 removal and process design data to full scale boilers. For example, current low-NO_x burner technologies appear applicable to only about 35% of the pre-NSPS coal-fired generating capacity. In addition, ash composition has been shown to have a major impact on SO_2 removal efficiency.

(3) Need for particulate control upgrading because of a 200% to 300% increase in fly ash loading and altered ash electrical properties.

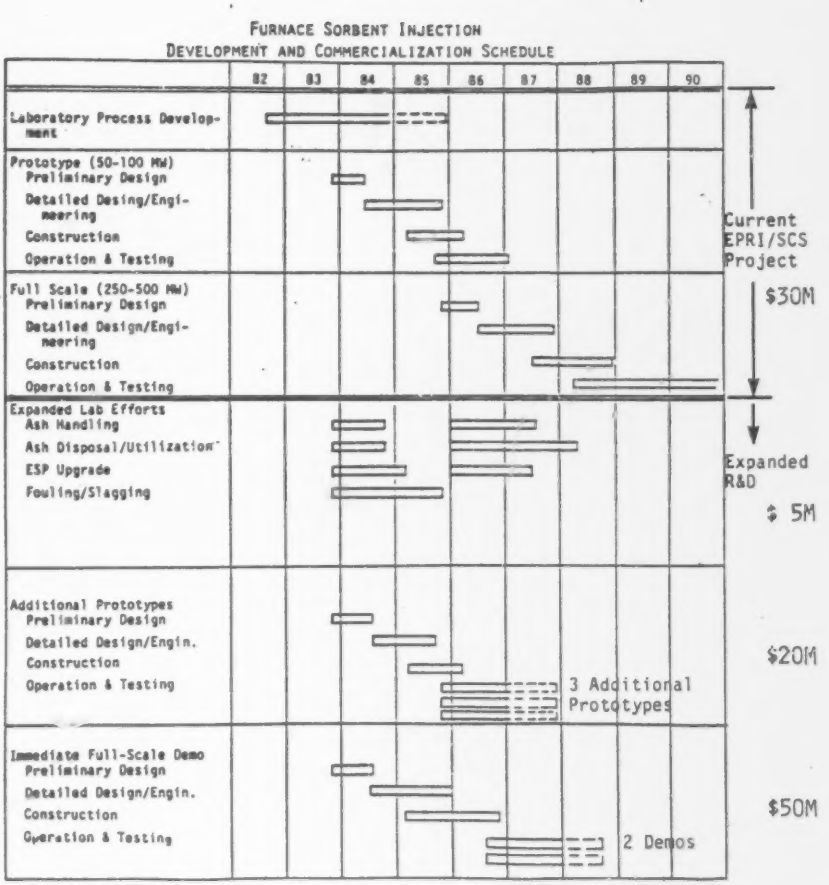
(4) Ash disposal constraints. Limestone injection could substantially increase the total ash quantities for disposal.

The goals of the various research programs, therefore, are similar but the specific research approaches are quite different, although complementary. In the U.S., development is principally supported by EPA and EPRI. To date research has focused on laboratory programs to both acquire the necessary process data and to evaluate power plant impacts prior to committing to commercial scale demonstrations.

These projects have established the groundwork for a major expansion of the EPRI research effort. This will include participation with utility companies in development testing, and prototype design and field evaluations during the next 3 years. EPRI participation in subsequent full scale utility demonstrations of the technology is also anticipated if warrented by development. During this same period, joint projects with the boiler manufacturers to develop and verify retrofit low-NO_x burners will also be completed.

The process development and commercialization schedule for high sulfur U.S. coal is shown in Figure 3. This schedule assumes a step-wise progression involving: (1) laboratory development ending in late 1984, (2) prototype verification testing of a small (50-100 MW) utility boiler in 1986, and (3) the first large (400+MW) utility boiler demonstration beginning in 1988. This step-wise approach will enable confident scale-up of the process from each stage in the development, thus providing a maximum probability of success and least risk to host boiler equipment. Interim results may permit application of the process earlier to some existing power

Figure 3


FURNACE SORBENT INJECTION
DEVELOPMENT AND COMMERCIALIZATION SCHEDULE
82
83
84
85
86
87
88
89
90
Laboratory Process Development
Prototype (50-100 MW)
Preliminary Design
Detailed Desing/Engineering
Construction
Operation & Testing
Full Scale (250-500 MW)
Preliminary Design
Detailed Design/Engineering
Construction
Operation & Testing
Current EPRI/SCS Project
$30M
Expanded Lab Efforts
Ash Handling
Ash Disposal/Utilization
ESP Upgrade
Fouling/Slagging
Expanded R&D
$ 5M
Additional Prototypes
Preliminary Design
Detailed Design/Engin.
Construction
Operation & Testing
3 Additional Prototypes
$20M
Immediate Full-Scale Demo
Preliminary Design
Detailed Design/Engin.
Construction
Operation & Testing
2 Demos
$50M

plants where process optimization is not required.

Because of the developmental status of the technology, no actual process cost data are available. However, retrofit cost estimates have been made for furnace limestone injection based on standard EPRI economic premises. The results indicate a probable capital cost in the range of $40 to 100/kW, including particulate control system upgrading and a total levelized cost of 6 to 12 mills/kWh for high-sulfur coal. This does not include the cost of any reduced boiler performance and increased maintenance resulting from application of this technology. This again is based on a SO_2 removal efficiency of 50% which leads to a cost per ton of SO_2 removed in the range of $650 to $1000. Thus the relatively low SO_2 removal efficiency and high limestone requirement may tend to offset the capital cost advantage. Based on required combustion conditions this option appears applicable only to at most about 35% of the candidate utility population or 70,000 MW.

WATER AND SOLIDS INTEGRATION

Little attention has been given to date to secondary pollution questions of acid rain mitigation, notably compliance with Clean Water Act/NPDES and RCRA. In addition to a very large increase in both solid and liquid waste products created by retrofit control requirements, scrubber chemistry will be different at lower removals. New approaches will produce byproducts of different characteristics--e.g., more soluble calcium in waste solids from furnace limestone injection than conventional wet limestone scrubbing.

In conjunction with planned EPRI programs for integration of the cooling tower with the SO_2 scrubber loop plus FGD chemistry, sludge disposal and groundwater protection, a full-scale integrated water treatment and solids management system at one or more existing plants should be constructed and tested. The system should focus on retrofit applications and be designed along with retrofit SO_2 controls. The effort would demonstrate application of retrofittable water quality control technology plus design and operating guidelines for retrofit water and solids systems.

ELECTROSTATIC PRECIPITATOR PERFORMANCE IMPROVEMENT

The retrofit application of in-furnace or certain other flue gas emission control technologies will have a detrimental effect on the performance and reliability of existing electrostatic precipitator (ESP) facilities at these plants. This degradation will result from both the increased particulate loading as well as the effects on fly ash resistivity, adhesivity, and particle size resulting rrom changes to fly ash and flue gas composition by the retrofitted control processes.

It will, therefore, be necessary to upgrade existing ESP technology including development of improved flue gas conditioning additivies, pulzed power supplies, on-line performance diagnostic instrumentation and improved fly ash properties prediction methods. This effort should include full-scale field evaluation of retrofit effects plus verification of performance improvements. The result will be utility proven techniques for maintaining and upgrading

ESP performance and reliability plus guidelines for the operation and maintenance of the upgraded ESP facilities responding to new SO_2 and NO_x control requirements.

FLUIDIZED BED COMBUSTION

The electric utility industry also is aggressively evaluating and developing technologies which may ultimately provide less costly and more reliable operation than current pulverized coal systems. These options are intended to carry utility coal utilization beyond the limits inherent in pulverized coal technology. The most important factor in commercial acceptance will be their demonstrated capability to be at least as reliable as pulverized coal-fired power plants. Accordingly, priority must be placed on reliability. This requires the construction and operation of large-scale, enginering prototype or pioneer plants over a range of designs and fuels at operating conditions which are representative of the using utility industry.

EPRI and the utility industry are, therefore, accelerating the development and application of fluidized bed combustion as a further evolutionary improvement in coal utilization to meet the expanding needs for coal-fired power generation. The primary goal is to reduce the cost of environmentally acceptable electric generation with coal and allow utilities to use lower grade fuels.

Development of atmospheric fluidized bed combustion (AFB) has successfully progressed from the process confirmation state to engineering prototype, making commercial utility scale demonstration practical. TVA has taken the lead, with EPRI support, in implementing a 20 MW engineering prototype which has been operating successfully for over one year. This facility has only been possible with major Federal R&D support through TVA.

The results provide the technical basis for 100-200 MW commercial scale demonstrations by the utility industry operational this decade. In support of this goal, EPRI has recently authorized $75 million for participation in a $220 million AFB demonstration cosponsored by TVA, Duke Power and the State of Kentucky. This was one of several utility AFB demonstration proposals submitted in response to an EPRI solicitation. Other such projects which we hope will proceed were submitted by Northern States Power, Colorado-Ute, Puget Sound Power and Light, and Cleveland Electric Illuminating Company. In each case the host utility proposed to fund the major portion of the cost of the demonstration, with EPRI and the boiler manufacturers sharing the risk costs with the utility.

AFB has developed to the point where it may also be a viable retrofit utility option to reduce SO_x and NO_x emissions as well as reduce sensitivity to coal quality. Of particular interest is the potential for conversion of existing pulverized coal boilers to AFB. The

confidence to achieve this potential will depend on successful demonstration such as that proposed to EPRI by Northern States Power. This project would convert in existing 115 MW boiler at a cost of less than $500/kW. The proposed startup date was October 1985. The capital cost may be partially offset by the resulting increase in generating capacity, fuel flexibility and usefully extended plant lifetime making it often a more cost-effective option than FGD.
Providing the necessary confidence in such a step puts a major burden on the technology development effort. If the private sector is to be responsible, then the demonstration becomes less the last step in the development process than an assured commercial venture. The continuity and success of the combined Federal/private development effort to date may make this pioneer demonstration step for AFB commercially viable primarily through private support. This, however, is likely to be the exception rather than the rule unless Federal support for coal combustion R&D, including critical pilot and demonstration plants, is maintained at a substantially greater level than currently authorized.

I am encouraged that the on-going Congressional review and mark-up of the requested DOE appropriation indicates possibility of augmented funding for the TVA/EPRI/Duke Power/Kentucky demonstration of AFB. If sustained, this support will be instrumental to completing the development and commercialization of an important utility coal technology whose success to date has resulted from the cooperative actions of government with the private sector.

SUMMARY

From the modification of the coal combustion process itself to the removal of SO_2 and NO_x from the flue gas, the possibilities for retrofit emission controls span a variety of technology alternatives. Depending on the degree and urgency of acid deposition control, the nation's pre-NSPS coal-fired power plants will be subject to quite different combinations of corrective action, even including the outright retirement of plants that do not justify retrofit.

For example, if the nation chooses to deal with the problem by requiring an arbitrary and uniform 8-12 million ton per day reduction in SO_2 emissions (as proposed by a variety of legislation), the control options become effectively limited to those currently available: plant retirements, low-sulfur coal blending, coal cleaning, and flue gas scrubbers. This approach would involve plants totaling about 200,000 MW of capacity. Making allowance for many kinds of uncertainty, it might cost $10-15 billion a year to execute, even if the various institutional and contractual limitations can be resolved. The large out-of-pocket costs imposed by this approach are primarily due to heavy reliance on scrubbers for more than half of this capacity. Another factor is the loss of generating capacity resulting from the power and reliability burden of these retrofit scrubbers and the retirement

of generating units that are unable to apply this option. This could reduce available generating capacity by more than 20,000 MW in the 1990's, a time when lack of new power plant construction may already restrict the availability of electricity to our economy.

Alternatively, strategies that allow for more gradual reductions in emissions would both open the range of retrofit control options and encourage the use of "least cost first" approaches. Retrofit options under development, such as furnace injection and low NO_x burners may provide somewhat more cost-effective emissions control but are probably limited in application and, therefore, have less potential for emission reduction.

Both of these scenarios offer something of a "band-aid" solution, and an expensive one at that. Besides the tremendous capital costs of add-on equipment or internal plant modifications, most of the options incur additional penalties in efficiency and/or reliability.

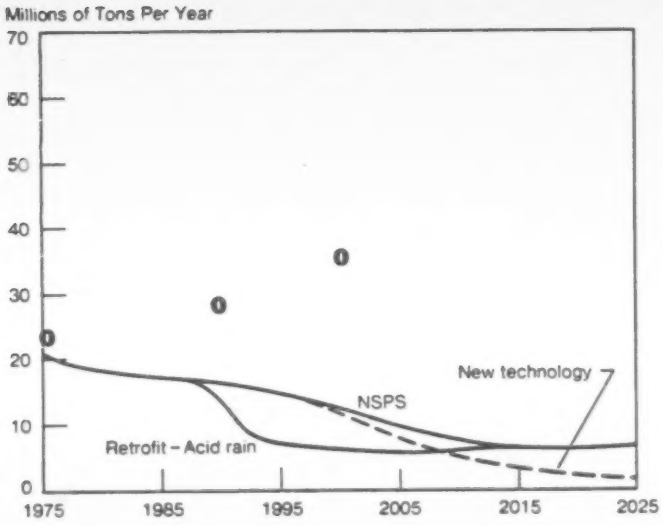
A perspective on the effect of reductions of utility SO_x emissions can be gained from Figure 4. Utility SO_x emissions have been decreasing for some time. For example, the decrease since 1975 is shown. If one projected a hypothetical uncontrolled case, the SO_x emissions could be as high as 35 million tons per year in 2000. But new units would be expected to meet the NSPS standards, and hence the emissions should follow the NSPS curve as indicated. If an 8 million ton per year reduction, aimed at reducing acidic deposition, were to occur, the SO_x emissions could decrease as shown between 1985-1990. This level of emissions continues for one or two decades and then rises to meet the NSPS case as the older retrofited plants built mostly before 1970 are phased out.

As new technologies, such as coal gasification combined-cycles and fluidized bed combustion, become demonstrated and commercialized, the total emissions could be reduced along the dotted pathway. This observation suggests the need for a balance between R&D support for technologies that can be adapted to retrofit applications and the new technologies that could be introduced to fundamentally resolve the perceived conflict between coal and the environment, while improving cost and efficiency.

EPRI has, therefore, given substantial R&D support to the development of intermediate Btu coal gasification-combined cycle (GCC) and fluidized bed combustion (FBC) systems. Both of these options show the promise of reducing SO_2 and NO_x emissions to very low levels. These are likely to be the effective coal based choices for the new generation of electricity beginning in the next decade.

The potential for these new types of generation is great enough to introduce another tradeoff consideration; although they are at least several years from commercial readiness, they allow emission reductions to follow naturally from the current economic and environmental evolution of coal-fired plants. This would elimiate

UTILITY SO_x EMISSION TRENDS



Based on EPA emission data and projections adjusted to reflect current utility growth projections.

- Basis average coal sulfur content — 1.8%
- Retrofit control for acidic deposition reduced SO_2/yr by 8 millions between 1985–1990
- Advanced technologies phased in after 1995

Figure 4.

costly band-aid approaches that serve to freeze technology in today's plants and in the long run would provide improved emissions control in a more cost-effective manner.

There is no shortage of opportunities to improve the efficiency, reliability and environmental perfomrnace of coal combustion. The successful achievement of the objectives laid out in this testimony requires, however, a more constructive and coordinated partnership between government and the private sector. This encompasses the planning and implementation of R&D as well as the commercial transfer of results. EPRI is prepared to do its part in formulating and implementing these efforts.

The actions of the Federal government in direct support of energy related research and development and in the form of incentives have a profound effect on the utility industry's ability to develop and use new coal utilization technology. Federal R&D policy with respect to the utility industry needs to consider the underlying circumstances affecting the industry and the importance of electric energy to the nation's energy, security and economic needs. The rate of commercialization and use of improved technology also depends on the development of regulatory incentives which encourage introduction of innovative technology. Too often our dependence on the adversarial approach to resolution makes the conflicts restricting coal use more severe and disruptive than necessary and restricts the introduction of technological improvement.

STATEMENT OF WILLIAM ELLISON, ELLISON CONSULTANTS

Mr. ELLISON. I would like to submit my 24-page prepared text to the committee. I would like, also, to read the conclusions of my effort, and then going over my notes, review the material—perhaps 25 minutes.

Mr. FUQUA. Can you make it a little shorter? We have some other witnesses.

Mr. ELLISON. I'll try to. Very good, sir.

Mr. FUQUA. It would be appreciated.

Also, without objection, a statement by Mr. Winn will follow the opening statement that I made at the beginning of the hearing.

Mr. Ellison?

Mr. ELLISON. Mr. Chairman, I was asked to make an overview of foreign technologies, principally but not exclusively in Japan and West Germany, as they might offer and where they might offer a helpful solution to some of the problems that Mr. Yeager has referred to.

The conclusions of my review, five in number, are as follows:

Worldwide SO_2 and NO_x emission control programs are technologically advanced and are a major source of alternative technologies for use in our future as well as current gas cleaning system applications.

Overseas technologies have already had important influences, as described, on major emission control facilities installed in the United States.

Any program for implementing backfit installations of flue gas cleaning facilities for SO_2 removal at existing powerplants will need to address, on a plant-by-plant basis, a host of potential site-specific constraints tending to prevent installation of a standardized, generically optimal FGD design, unilaterally.

Four, the diversity of site conditions and constraints plant by plant should provide a significant opportunity and justification at

each candidate site to assess and choose from candidate foreign, and United States, design approaches such as those I will review briefly today.

Significant opportunities, some detailed herein, exist for governmental actions apart from the contemplated acid rain legislation, to help insure that the design, operation, and reliability of SO_2 and NO_x control facilities meet acceptable standards and further environmental pollution control objectives in a cost-effective manner.

To give as many of the details of my review as possibie within the allotted time, I'll go through my notes very quickly and attempt to give you the high points of my findings and my knowledge on the subject.

As indicated, the technology development abroad has been impressive. The Japanese program, motivated by and emphasized by ambient air quality standards that are substantially more stringent than in the United States, and with a population density and other factors, have moved very dramatically, as most people know, in this area, and there's a great deal to be said about the benefits we've enjoyed already and the future possibilities in use of Japanese technology.

This technology has, in several instances, had a dramatic effect on the progress of our growth in knowledge in this country. In that regard, I'm referring both to the now generally common use of forced oxidation systems as well as the application and the very promising future in use of dry scrubbing, which is largely, but not entirely, a byproduct of a highly skilled, industrial spray drying technology from, of all places, Denmark.

There has been a dichotomy in our adoption, through a bold and progressive program by EPA and TVA—there's been a bit of a dichotomy in the way in which we've taken the forced oxidation technology applied in Japan to, in our own inimitable manner, come up with a throwaway waste disposal process. I'll come to more details on that in a moment.

There are a host of constraints that I will discuss in just a moment that make each site an individual problem, as it were.

The specific examples of governmental action, which in my view, as a practicing engineer, would greatly help this program, would be steps that would be taken through EPA and other agencies to better insure that we will monitor, perhaps participate in, and have full cognizance of these many developments, both in Japan and West Germany and make the fullest utilization of them in such a manner as to minimize the types of technical development in this country that would be seen to be paralleling these on a do-it-yourself basis.

Second, we have from Japan an outstanding example of a way in which, through a system of national licensing of air pollution control system operators, we could bring to bear the greatest amount of training, professional and societal rewards, and respect for the job of operating these systems, as has been the magnificently successful case in Japan.

Third and last, as part of a major concern in regard to the disposal of wastes from the many systems that the Congress is contemplating—calling for—there is a need for a use of systems that

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can—economically justifiable systems that can—yield a byproduct material that can be used in our economy.

We have some spectacular programs within the DOE for development of processes such as Eberra from Japan, Sulf-X from Pittsburgh, Pa., that will in due time bring us combined SO_2-NO_x technologies with valuable byproducts that will be attractive, will probably be quite attractive in many applications. Time is against us.

We have at the present time and for the short range a need for the fullest opportunity to pursue the use of byproduct systems as is universally used in Japan and West Germany, and there's an important hindrance to that that you could do a great deal about, and I'm referring at this point to a system, the system of depletion allowances in the United States in the IRS Code, in which we do a good job of insuring that our domestic and foreign raw material resources will not be depleted, but instead will be renewed by new discovery.

In the area of gypsum, we, as in many other substances or materials, raw materials, we have depletion allowance, and it's the considered opinion of many workers in the field, including myself, that the gypsum depletion allowance is of sufficient magnitude to, for all intents and purposes, prevent the ready marketability of potential byproduct gypsum sources in this country and to discourage and to serve as a disincentive for a broader use of byproduct systems, as has been the case in foreign countries.

I believe this could be done in such a way as to make the U.S. gypsum industry the hero, rather than the goat, of our solid waste problems in this country, in such a manner as to provide for the same granting of allowances in IRS tax reporting for by product gypsum utilization as now prevails in the case of natural gypsum deposits owned by the manufacturer.

This could conceivably be done by a hypothetical assumption that the gypsum was derived from an existing mine, so that without injuring, but rather helping, the U.S. gypsum industry to help us in our air pollution and solid waste management problems, we will allow them to have now the additional benefit, beyond what they already have, of an incentive to vigorously in a free market circumstance negotiate with utility companies so inclined to produce and use a byproduct gypsum. This use of gypsum has been largely motivated, and entirely motivated in systems of that type to date in this country, by the shortage of available land and not by any altruistic attitude, but rather a pragmatic need to find a way of moving the material out of sight and out of mind.

There will continue to be, in my opinion, many such opportunities, but those opportunities will not be realized unless we can remove the disincentive for the use of the very good quality byproduct gypsum by the many gypsum wallboard plants in the United States.

The foreign technologies from which we've drawn have been the use of the forced oxidation in Japan. We've done a good job in the last 7 years of bringing that same technology along with all of the aspects of it that in Japan, and now in Germany, and hopefully here in the United States, will bring us better availability, better reliability in our high-sulfur FGD systems, our present circum-

stances being such as to provide reliabilities that are not cost effective and are not satisfactory.

The advent of the dry scrubbing technology, largely, as I said before, a foreign technology brought here, and now extremely well received by the utility, involves the scrubbing of the gas in a device called a spray dryer, the same kind of device in which you might make instant coffee. And it has some outstanding advantages and is now being dramatically—moved into the market.

We've had, indeed, a spectacular, in my view, a spectacular, activity within DOE itself in terms of the fine program that has been instituted at the Argonne National Laboratories in equipping the small industrial boiler there with a—burning Illinois coal, a high sulfur, and equipping it with dry scrubbing. There are some outstanding results that are going to be reported later this year at the EPA-EPRI symposium attesting to the efficacy of the systems on high-sulfur coal, and particularly establishing an extremely novel feature of dry scrubbing not generally known and realized by the average worker in the field, that whereas wet scrubbers are ineffective in SO_3 removal, a small tramp sister to SO_2, which SO_3 in high-sulfur applications creates considerable corrosion havoc, the dry scrubbing system has now been shown and published data appears clearly to show that it, irrespective of sulfur level in coal, brings the sulfuric acid dew-point temperature down to the same uniformly low, benign level and seems to offer one important opportunity, one important gift in terms of dealing with the poor availability on high sulfur.

Mr. Chairman, how much time do I have left?

Mr. FUQUA. We still have some questions, if you can maybe in 5 minutes——

Mr. ELLISON. Yes, sir, thank you.

I believe Congress time would be best spent in closely examining the landmark legislation that was passed this year in Germany, which I'm sure many of you know about. I'm referring now to some of the details of this legislation which, in a very dramatic fashion, has set up a program in Germany in which, over the next 5 years, all existing units in excess of 300 megawatts will either be retrofitted or will be phased out of existence. Similarly, those smaller units, below 300, will do the same thing in 10 years.

I believe there is within the framework of this work that they have done and these rules that they've set up a possible basis on which Congress could grapple with the matter of the old boiler, with only a 5- or 10-year remaining life, in terms of the way in which, either through the incorporation of LIMB or some other low capital cost technology, those boilers might be permitted to be phased out gradually, less gradually in the case of those boilers which elect to use some means such as LIMB, high operating costs as it might be.

There are, indeed, some rather important new work on LIMB going on. I don't hear much about it in the States, and I'm not clear how well it's understood by the average worker. In Berlin a 700-megawatt boiler burning a coal very similar to our bituminous coal will be in operation, fully retrofitted with LIMB, using lime rather than limestone, by sometime next year, and it appears to me that we have the prospect through the vigilant monitoring of

this work overseas that we might have an early answer on the matter of the use of LIMB, which could be very valuable in keeping some of our boilers going to their proper final lifetime.

The users—the developers of the LIMB on the bituminous-like coal forecast that it will be a natural for small boilers and that on old boilers it will, regardless of size, that it will be a cost-effective way of control.

My last point has to do with the matter of the dichotomy in our progress in the use of the European and Japanese technology to date. I consider it rather significant oversight on the part of many, or most, or all, designers of FGD systems in our attempts to use forced oxidation as the Japanese have and gain all the advantages that we are anticipating.

We are winding up with a method of solid waste management which is far less effective, and will be far less effective, than older methods in the early seventies here in controlling impacts on fresh water receiving streams, so much so that the support area of technology pertaining to the treatment and disposal of saline wastes that is being sadly neglected as we proceed with many of the projects that are going on.

There are 35,000 megawatts worth of forced oxidation FGD systems that have been committed to date, and many of them in recent years.

There is, particularly in Germany, an important emphasis that I think we can well afford to pick up in the use of regenerative methods of use of the 300-degree raw flue gas to serve many purposes, one of which would be to convert to a byproduct salt cake these saline wastes, so that we would not impact our surface water streams with these methods of drying gypsum, so that it would be more salable; methods of agglomerating gypsum, so we could be more salable. So there are many little items on the menu as we look over these foreign countries.

And my very last point, Mr. Chairman, the highest of credit to EPA for an extremely magnificent program in Japan in monitoring the work in Japan, as they have for many years, through Dr. Andro of Chuo University, and they have been very effective in bringing to the attention of everyone what's over there and what's possible. We need an equally—equal zeal now that Germany's program has led to in excess of 10,000 megawatts of FGD systems, but we need an equal zeal in now proceeding to in Germany at least as much as we've done so far as surveillance in Japan. We need a Dr. Andro to come out of the wings in Germany to serve a comparable purpose.

I thank you for this opportunity to speak to you today.

[The prepared statement of Mr. Ellison follows:]

OVERVIEW OF WORLDWIDE TECHNOLOGIES FOR REDUCTION OF EXISTING STATIONARY EMISSIONS OF SO_2 AND NO_x

William Ellison PE
Ellison Consultants

Written Testimony to Hearing on
"Acid Rain: Implications for Fossil Energy R & D"

SUBCOMMITTEE ON ENERGY DEVELOPMENT AND APPLICATIONS

and

SUBCOMMITTEE ON NATURAL RESOURCES, AGRICULTURE RESEARCH AND ENVIRONMENT

of

U.S. HOUSE OF REPRESENTATIVES COMMITTEE ON SCIENCE AND TECHNOLOGY

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SUMMARY
of Written Statement of WILLIAM ELLISON PE

Overview of Worldwide Technologies for Reduction
of Existing Stationary Emissions of SO_2 and NO_x
to
Hearing on "Acid Rain: Implications for Fossil Energy R & D"

The impressive development and application of principal SO_2 and NO_x control technologies by Japanese and European firms and industries is described to provide a background for understanding identified ways in which many of these technologies have already contributed to the implementation of acid gas emission control programs that have previously begun in U.S.A. Details are given of a major dichotomy tied to widespread adoption in U.S.A. of forced-oxidation flue gas desulfurization (FGD) concepts from Japan; this technology transfer has led to a new unique-to-U.S.A. method(s) of throwaway disposal of gypsum-like FGD solid waste that retreats from a previous evolving U.S. FGD solid-waste disposal practice that appears to better control potential impact of FGD liquid waste on natural groundwater and surface water bodies near the throwaway disposal site.

Significant overseas technologies for implementing a new U.S. program to reduce emissions from large existing sources are identified and described in light of the many potential generic constraints that, one or more, could hinder the cost-effective design and operation of efficient and reliable control systems at individual sites.

Pointed examples are offered of needed governmental actions to ensure best utilization and application of relevant existing and emerging worldwide technologies in reducing stationary emissions of SO_2 and NO_x. These pertain to the clear value of improved means of ensuring fullest utilization of pertinent foreign technologies; the need for a program of nationwide scope to upgrade gas cleaning system operator effectiveness and to help bring up FGD system reliability experience to a cost-effective level; and to the advantage of adjusting IRS gypsum depletion-allowance tax provisions to reduce the disincentive for private industry's use of large outputs of potentially available FGD by-product gypsum; if possible, without financially penalizing the U.S. gypsum industry because of perceived, potentially significant, national FGD solid waste management problems.

CONCLUSIONS

This overview indicates that:

1. Worldwide SO_2 and NO_x emission control programs are technologically advanced and a major source of alternative technologies for use in our future as well as current gas cleaning system applications.

2. Overseas technologies have already had important influences, as described, on major emission control facilities installed in U.S.A.

3. Any program for implementing backfit installation of flue gas cleaning facilities for SO_2 removal at existing power plants will need to address, on a plant-by-plant basis, a host of potential site-specific constraints tending to prevent installation of a standardized, generically optimal FGD design.

4. The diversity of site conditions and constraints plant-by-plant should provide a significant opportunity and justification at each candidate site to assess and choose from candidate foreign (and U.S.) design approaches such as those reviewed herein.

5. Significant opportunities, some detailed herein, exist for governmental actions apart from contemplated acid rain legislation, to help ensure that the design, operation, and reliability of SO_2 and NO_x control facilities meet acceptable standards and further environmental pollution control objectives in a cost-effective manner.

I. INTRODUCTION

This assessment of present and emerging overseas technologies for SO_2 and NO_x reduction is focused on flue gas cleaning and other control techniques that may lend themselves to retrofit (backfit) application on a number of large existing bituminous coal fired boilers in eastern United States. Particular attention is given to potential generic constraints on flue gas treatment backfits at such candidate sites and possible means of optimizing retrofit designs based in part or in entirety on technologies, methods and practices of foreign origin. An outline is offered of relevant government actions in U.S.A. that may help achieve a successful plant-level outcome in the operation of facilities installed in conjunction with any backfit emission control program to be adopted in U.S.A.

II. FOREIGN STATIONARY-SOURCE CONTROL PROGRAMS

A. GENERAL

Significant system technology for control of sulfur dioxide and, to some extent, nitrogen oxide emissions has been developed and applied in Japan, earliest installations being primarily for residual oil fired boilers, the most recent on new coal-fired units. Its program, like that of U.S.A. for SO_2 control, began in more than ten years ago with major flue gas treatment installations coming on line in 1971.(1) Strong acid-rain precursor control programs incorporating stack gas cleaning on coal-fired boilers have begun in Europe more recently, making substantial use of Japanese and U.S.-developed technologies as well as system designs originating in West Germany.

B. IN INDIVIDUAL COUNTRIES

Worldwide foreign nations significantly involved in generation and some degree of abatement of the acid gases from stationary sources may be considered to constitute 4 regions: Japan, Mainland Europe, England and Canada.

(1) Japan

In assessment of the Japanese work, recent extensive application and commissioning of SO_2 and NO_x removal facilities for coal-fired boilers is of particular interest, (initial startup beginning in 1976 and 1981, respectively.)(2) This impressive new effort (3) is tied to ambient air quality standards for SO_2 (approximately 0.02 ppm yearly average) and NO_x (0.02 to 0.03 ppm yearly average) that are substantially tighter than U.S. criteria. It comprises backfits and new plant installations in conjunction with a national program to increase coal-fired power generation to 22,000 MW (10% of total utility power) in 1990 as contrasted with 4,300 MW (3.7%) in 1979.

a. Principal Technologies Used-- Many innovative and potentially attractive control technologies have been applied and developed in Japan. Of key significance, however, is the above noted sequence of retrofits and new-plant installations for coal-firing, which very commonly control acid gas emission by an in-series arrangement of:

- Ammonia-based selective catalytic reduction for dry NO_x collection upstream of the boiler air preheater in the amount of approximately 80% removal using a large honeycomb catalyst bed and
- limestone-based forced-oxidation flue gas desulfurization (FGD), in which wet scrubbers achieve in excess of 90% SO_2 removal, converting it to usable gypsum (double-hydrated calcium sulfate) by-product, and treating and outboarding, generally to a nearby sea water body, FGD saline liquid (liquor) effluent containing dissolved calcium chloride originating principally from hydrogen chloride acid-gas scrubbed out of the raw flue gas. (Electrostatic precipitators for particulate emission control, installed upstream of FGD, limit the amount of tramp

ash entering and collected by the FGD system and intermixed with the gypsum by-product.)

Since the beginning of Japan's control programs this FGD method has been the predominant one in Japan, both for residual oil and coal fired boilers.

b. Significance of Use of Forced Oxidation-- The use of forced oxidation, (introduction of compressed air into the scrubbing slurry, usually at the point of lowest pH in the system,) in Japan usually in an external air-slurry contact tower, has aptly served the following important purposes:

- Supply oxygen to convert the absorbed SO_2 to sulfate, rather than to sulfite that is yielded by SO_2 absorption when oxidation is absent.
- Provide fine suspended gypsum seedlings in the recirculating scrubbing slurry so as to provide suitable sites for the orderly precipitation of the SO_2-catch as gypsum and prevent scaling of the system internals.
- Improve process operation by maximizing limestone utilization, SO_2 removal efficiency, etc.
- Ultimately render the SO_2-catch as a comparatively coarse-grained gypsum mineral, which, after suitable fresh-water washing, is of a chemical quality suitable for all major commercial gypsum uses including manufacture of wallboard.
- In conjunction with an ample blowdown of FGD liquor to reduce and limit its dissolved solids concentration it affords a stable and well-controlled FGD chemical-environment yielding extremely high system reliability e.g. 98%.

c. Extent of By-Product Gypsum Production-- The current annual output of FGD by-product gypsum, sold at anaverage price of 3,000 yen/metric ton (approximately $10/short ton), is approximately 3 million tons, which is supplied to the domestic market totalling over 6 million annual tons. FGD gypsum revenues are equivalent to only about 5% of the annualized FGD cost. The principal benefit of the by-product mode is elimination of need for land space for storage/disposal of the SO_2-catch [4].

(2) Mainland Europe

Coal-burning countries of mainland Europe have begun major programs for acid gas emission control.

a. West Germany-- The most recent (June 14, 1983) update of the Federal Air Pollution Control Act requires installation of FGD units within a maximum of five years on all existing coal-fired utility units larger than 300 MW, ten years for smaller units. To date approximately 10,000 MW of time-staged FGD modules for twenty coal-fired units have been ordered and/or installed with initial startup date of FGD modules ranging from 1977 to 1988 [5]. A large proportion of them are retrofits. Total annual gypsum production from these operating and committed units will be approximately 2 million short tons. State-of-the-art related NO_x emission standards have been established with the aim of bringing about use of selective catalytic reduction. Approximately one half of the FGD

installations are of German designs, the balance being of Japanese or U.S. origin. But with the exception of one unit all are designed with forced-oxidation to produce salable gypsum and in the case of at least one, FGD gypsum is presently being used for wallboard manufacture. Thus, SO_2 control technology being used in the relatively new West German program is generally similar or comparable to the recent installations in Japan.

b. The Netherlands- Following closely the FGD approach in West Germany, by-product gypsum yielding forced-oxidation scrubbing systems have to date [6] been purchased for two 300 MW retrofits on existing coal-fired boilers from among a population of 2,100 MW of existing coal-fired capability in The Netherlands. An additional 1,400 MW of gas/oil fired utility capacity currently being converted to coal will also be equipped in this same manner and provision for space will be made to accommodate selective catalytic reduction [7]. Annual gypsum production from these committed and planned systems will total approximately 0.5 million short tons.

c. Other Countries-- In Sweden two coal-fired plants have recently been equipped with throwaway-waste spray-dryer type dry scrubbing systems [8]. In Norway no coal is used in power generation, but seawater-scrubbing FGD systems have been installed on residual oil fired boilers at a large chemical plant and at an oil refinery, and pending at large aluminum plants [9].

(3) England [10]

No significant technology is in use or pending. Although the U.K. Government has ratified the EEC directive to take positive action to control the emissions of SO_2, no retrofit FGD is planned. The nationally owned power company, CEGB, acknowledges that future large fossil-fired units will need some type of FGD, but no projects are active because there will be a nationwide surplus of generating capacity at least until the late 1980s.

(4) Canada

An initial stack gas cleaning program is expected to focus on existing high-sulfur coal fired boilers in the Atlantic maritime provinces [11]. Commercial FGD processes known to be of greatest current interest to the utility industry are throwaway-waste type including seawater scrubbing developed in Norway.

III. PRINCIPAL INFLUENCE OF FOREIGN TECHNOLOGY IN U.S. CONTROL APPLICATIONS TO DATE

Throwaway-solid-waste type FGD using lime or limestone reagent has continued to provide a less costly control method than by-product systems typically used overseas, and it remains the most common scheme for coal-fired units in U.S.A. However, beginning in the late 1970s, foreign technology has had a pronounced effect in revolutionizing our design practices for throwaway-waste FGD.

A. LIME/LIMESTONE BASED WET SCRUBBING

(1) Earliest U.S. FGD Technology Base

Up to the late 1970s most U.S. utility FGD systems purchased were natural-oxidation type lime/limestone wet scrubbing operations. This process, normally installed downstream of electrostatic precipitators that control particulate emissions, yields a sludge-type solid waste that is fine-grained, highly water retentive and thixotropic, (i.e. tending to become fluid when mechanically disturbed.) Because of potential environmental problems in land-based ultimate disposal of this voluminous waste and the inability to reclaim land inundated with the material, many such plants adopted a method of sludge fixation (stabilization) that consists of chemical processing of scrubber sludge to facilitate handling, transportation, placement, and consolidation at the ultimate disposal site. One of the methods used is pug-mill blending of mixtures of FGD filter cake, dry fly ash, and other dry additives, followed by landfill compaction. An alternative procedure involves the addition of silicate-containing materials to thickened FGD slurry before discharge to a pond site. Extensive testing indicates that these methods of chemical treatment significantly improve the load-bearing characteristics of FGD sludge, reduce the solubility of the major chemical species by a factor of 2 to 4, and reduce sludge permeability by an order of magnitude or more. Further, the encapsulation of a substantial amount of FGD liquor in the consolidated waste mass, approximately one part by weight per part of raw-sludge solids, provides the option for environmentally acceptable, all-in-one, combined disposal of all solid and liquid waste without discharge of a liquid outfall.

(2) Supplanting of Natural-Oxidation FGD by Forced-Oxidation FGD

Based on the merits of forced-oxidation (FO) FGD as demonstrated overseas, notably in Japan, its advantageous adaptation and use for throwaway-solid-waste operations in U.S.A. were recognized by about 1976 by the U.S. EPA and the electric utility industry. Since that time approximately 20 power plants constituting 16,000 MW of capacity have purchased FO-FGD (or made provisions to backfit FO in natural-oxidation installations). This represents half of all limestone FGD committed to date in U.S.A. As noted earlier, (Section II B 1), this SO_2 removal method is superior to natural-oxidation FGD commonly applied in U.S.A. since it provides improved control of FGD-process operating conditions serving as potential basis for higher FGD availability in limestone based

SO_2 absorption. It results in improved control of internal scaling, better utilization of limestone chemical reagent and enhanced SO_2 removal efficiency. Moreover, the solid waste produced from the SO_2-catch is in the form of comparatively coarse-grained calcium sulfate solids that may be more easily transported and stored and which could, if desired, be rendered in the form of a usable gypsum by-product. However, only for 13% of the U.S. FO-FGD capacity committed to date has the utility company elected to design to yield a usable gypsum by-product,[12] the annual gypsum yield from these few totalling only about 1 million short tons. Thus U.S. adoption of the Japanese forced-oxidation technique has led designers to a unique and now commonly applied throwaway-mode of solid waste management.

(3) Waste Management in FO-FGD Installations

Throwaway FO operation generally yields typical gypsum crystal size of from 40 to 100 microns, producing an easily dewatered and handled coarse-grained cake, which can, if necessary, be readily transported as an 80-90% solids cake to a remote disposal site. Moreover, studies have shown that the acre-feet solid-waste disposal capacity required in conjunction with an FO-FGD waste-landfill operation is less than 60% of that for direct ponding of sludge without forced oxidation. Of course, fly-ash stabilized sludge formed at many plants requires measureably more space for this mixture than for the sludge alone. Based on field tests [13] a major simplification of FO-FGD for solid waste dewatering and ultimate disposal may be anticipated; it has been demonstrated that FO-FGD solid waste may instead be discarded by wet stacking, a technique for land-based disposal of waste-gypsum, utilized extensively in the phosphoric acid industry in Florida, wherein a 10 to 15% solids aqueous-slurry is discharged at a disposal site so as to permit formation of a sharply-pitched stack over the plant lifetime. In this method now being planned for a number of power plants, and of strong interest in Canada as well, reduced acreage use is achieved through such stacked disposal without need for dewatering by thickeners and filters or centrifuges. Overall capital costs and annual revenue requirements are understood to compare favorably with even simple ponded-waste natural oxidation FGD systems. In the so-called "upstream method" of construction the wet stacking is begun by first constructing an earthen starter-dike forming a sedimentation pond. FO-FGD solid waste is pumped to the pond in a slurry form, usually at 10 to 20 percent solids, and allowed to settle and drain.[14] Process water is decanted from the pond and returned to the plant. When sufficient gypsum is deposited within the pond, gypsum is excavated with a dragline to form and step-wise build up a perimeter dike immediately inside the starter dike. Decanted liquor is piped under the starter dike to an annular perimeter-ditch and surge pond formed by the starter dike and a clay exterior dike. The process of sedimentation, excavation, and raising of the perimeter dikes continues on a regular basis during the active life of the stack. Using the upstream method of construction, gypsum stacks in the phosphate industry have reached heights exceeding 100 feet (30.5 m) with slopes of 1.5 horizontal to 1.0 vertical, which is approximately the angle of repose of some

gypsums. These steep slopes result from casting the gypsum with a dragline and allowing some gypsum to roll down the outside of the stack to eliminate need for shaping it.

(4) Dichotomy in FO-FGD Waste Management Practices

Throwaway-solid-waste FO-FGD operation based on U.S. designs is a marked reversal of earliest FGD design strategy that was typically and logically based on exercising the option to limit/avoid outfall discharges of saline FGD wastewater by the purging from the system of a substantial amount of surface moisture (liquor) occluded in the fine-grained natural-oxidation sludge. FO operation so drastically reduces waste-solids surface moisture content, e.g. to 10% by weight, and so little of the liquor can be retained in the discarded coarse-grained waste mass over an extended period, that the closed-loop FGD criterion appears clearly to be no longer valid. This is because of the major stepwise increase in system corrosion phenomenon and in process design complexity that would accompany an approximately 10-fold or greater increase in FGD-liquor concentrating effects. However, evaluation, planning and design of throwaway-waste-FGD systems in U.S.A. continues to be predicated in almost every case on closed-loop operation; and on the erroneous assumption that surface moisture occluded in the FO-FGD waste, little though it may be, will, as in the case of the thixotropic natural-oxidation sludge, be retained for the long term and serve as a finite liquid blowdown from the system. This misconception and the assumption of closed-loop operation is of equal or greater concern in any application of wet-stacking since leachate flow through the stack mass will recycle in an uncontrolled manner and be returned again to the recirculating scrubbing liquor stream. Moreover, liquid waste management may be expected to be a major problem in wet-stacking operations since the steep slopes of the gypsum perimeter dikes will have a factor of safety very close to unity and, from a conventional geotechnical engineering point of view, gypsum stack failures will occasionally occur. However, gypsum is a very forgiving material, and, unlike most mine tailings, gypsum does not readily flow. Therefore, the consequence of a failure is the loss of process water stored on the gypsum stack. If process water escapes the plant property, liabilities from pollution and environmental damage may also result. On a day-in, day-out basis, seepage of process liquid to groundwater and surface water would need to be controlled by use of an impervious foundation as well as of clay-type exterior and starter dikes, and by collecting and recycling, or treating and outboarding all seepage.

Environmental concerns in use of wet-stacking for FO-FGD waste include:

- The very large working area of the stack facility will cause the liquor recycle to FGD (decanted from the active-pond section) to also return large quantites of runoff liquid. Resulting water balance problems in rainy weather will lead to intermittant outboarding of substantial process-liquor discharges.

- Although stack erosion and creation of windblown dust is claimed not to be a significant problem, all pile run-off will be saturated with calcium sulfate and contain cake surface-moisture constituents.
- Over a twenty-year lifetime a 100 feet high stack will be created. In no country are regulations or experience available as to how to satisfactorily decommission a lifetime gypsum stack or to reclaim it.
- Stack slopes are too great to hold topsoil and vegetation. Thus, normal concepts for "covering" of the waste in this manner do not apply.
- The pile must be free draining or it will lose stability [15]. After the FGD lifetime, seepage of leachate will continue unabated with no user to which to recycle. The substantial solubility-in-water of the gypsum solids will yield an endless source of leachate saturated with calcium sulfate plus other components such as sulfite (COD) plus any remaining surface moisture constituents.
- Cold winters will severely restrict dragline operations during cold-weather months and jeopardize continuity of FGD operation. In addition freeze-thaw action is of concern so far as maintaining the integrity of dikes.

B. LIME BASED DRY SCRUBBING

Due principally to advanced industrial spray dryer technology in Denmark and its introduction and application in the U.S. through U.S.-based sales operations of the Danes, major use has been made of dry scrubbers of the lime spray drying type in U.S.A. beginning in 1978[16]. To data approximately 6,500 MW of lime spray drying FGD, a throwaway waste process, have been purchased in U.S.A., more than 70% of it from Danish firms. Although these are primarily low-sulfur western U.S. coal-applications, test runs have been carried out on industrial and small utility-plant size installations fired with high sulfur coal [17]. Extensive utility plant application in high-sulfur coal service appears to be possible. The principal incentives for use of lime-based dry scrubbing in lieu of wet limestone FGD have been:

- Adequate SO_2 removal without water saturating the scrubbed flue gas, thus circumverting the corrosive low-pH type wet corrosion tendencies in the flue-gas circuit downstreams of the mist eliminators of wet scrubbers, (the section that causes the greatest impairment of reliability for this latter type of FGD)
- Absence of need for recirculating slurry system and solid waste dewatering facilities
- Generally lower capital cost
- Lends itself to throwaway management of solid waste with minimum of difficulty caused by wetness/moisture in alternative wet scrubbing applications.

IV. MAJOR SITE-SPECIFIC CONSTRAINTS IN CONTEMPLATED BACKFIT OF CONTROLS TO EXISTING U.S. UNITS

Any existing unit and site to be backfitted with FGD may have one or more limitations that would prevent installation of an optimal design. Constraints as follows will be of significance in weighing alternative technologies and in identifying and assessing technologies, methods and practices, of foreign origin in this instance, that may best address potential retrofitting problems.

A. PLOT SPACE FOR CONTROL EQUIPMENT

Adequate area will be required immediately adjacent and downstream of existing electrostatic precipitators and/or the plant stack to accomplish a practical re-routing and connection of flue-gas ductwork. In the worst-case scenario, lacking substantial room, the simplest single-module high-gas-velocity wet scrubber module may be the only feasible system installation.

B. REMAINING PLANT-LIFETIME

If a very old unit is to be controlled, the remaining lifetime of the unit may be so short, e.g. 5 to 10 years, as to preclude expenditure of typical capital investment required to minimize operating costs. This circumstance could lead to an economic decision to meet SO_2 reduction objectives by use of a cleaner fuel source and/or fuel cleaning in addition to the lowest-capital-cost FGD method that will meet the balance of SO_2 removal required.

C. PERSONNEL

An old plant may not have or be able to successfully recruit and/or train manpower to provide the necessary skills in operating and maintaining a process-type system operation. Such a constraint would most logically occur/exist in a circumstance in which many FGD systems were being installed in the region.

D. COAL SULFUR LEVEL

As contrasted with Japanese and European plants, eastern U.S. electric utility units fire coal fuel with very high sulfur, e.g. 3 to 5% by weight. Experience in the U.S.[18] indicates that FGD reliability, (the accumulative amount of time the system was operated during any calendar period divided by the duration of time periods during which the system was called upon to operate) has consistently averaged, nationwide, only approximately 81 to 82%, and in the case of high-sulfur installations only, is substantially lower. Thus, high-sulfur coal service is a difficult application due, in part, to intensified corrosion effects in internals downstream of the scrubbers exposed to low pH condensate from wet gas, calling for use of technologies methods and practices that offer a basis for improved system reliability in U.S. installations.

E. ACCESS TO WASTE DISPOSAL SPACE

It may not be feasible in or near the power plant to dedicate large tracts of land required for disposal of FGD solid wastes over the lifetime of the FGD-equipped unit(s). In this event a system design to yield a salable solid waste by-product would be of great interest.

F. EXISTING FLY ASH COLLECTION FACILITIES

If existing electrostatic precipitators are not performing reliably, operation of a wet-scrubber type FGD system-selection could be adversely affected due to the effect on system chemistry of tramp components of fly-ash collected in the scrubber and/or the fly-ash adulteration of the FGD solid waste in a possible application calling for usable by-product.

G. RECEIVING STREAMS FOR LIQUID EFFLUENT

Many inland power plants in U.S. are sited on rivers that may be adversely impacted by potential saline liquid effluent from new FGD installations. Where this is the case, selection of wet scrubbing processes requiring open-loop operation may be prevented or special design provisions would be needed to achieve ultimate disposal of the dissolved solids loading of the required blowdown.

H. EXISTING STACK

It may be necessary/appropriate to retain the existing stack for use with the FGD-equipped unit(s), and the stack design/construction may not be suitable for use in cool/wet flue gas service. Provisions for substantial reheating of cleaned flue gas may be called for to protect the stack.

V. OVERSEAS TECHNOLOGIES AND PRACTICES OF SIGNIFICANCE IN PENDING U.S. BACKFIT PROGRAM

Many recent Japanese and European technological advances and commercial developments may be utilized to minimize backfit problems and annual cost, help ensure successful performance and maximize reliability of new retrofits in U.S.A.

A. WET SCRUBBING

It may be expected that commercial improvements that continue to be made in overseas FO-FGD system designs and practices will provide means of improving the tailoring of such FGD systems for existing U.S.plants. A number of the candidates may be grouped as follows:

(1) Plot Space Related

Where applicable at individual sites due to space constraints and/or to achieve cost economies, it will be possible to use large single-module absorbers of 350MW and greater such as have been developed and utilized in both West Germany and Japan. Small diameter and large gas flow capacity is particularly emphasized in available and commercially demonstrated co-current absorber towers operating at elevated design gas superficial-velocities, e.g. in excess of 20 feet/second.

(2) Personnel

As discussed further in Section VI, a recent U.S. EPA field tour and survey of new commercial stack gas cleaning installations in Japan has determined that a major national program for training/schooling and certification of pollution control system operators during the past ten years has had a very significant impact in improving the competence and performance of plant personnel and in maximizing gas cleaning system reliability throughout that country. Support by the utility industry for establishment of such a program in U.S.A. would help to upgrade operation of acid gas control facilities and ensure effectiveness of capital investments required for their installation.

(3) High Sulfur Coal

A possible conclusion to be drawn from the very wide disparity in reliability experience, U.S.A. on high-sulfur coal versus overseas applications on lower-sulfur coal, is that impairment of U.S. operations is caused to a great degree by increased flue-gas SO_3 (and SO_2) concentrations tied to elevated sulfur levels in most Eastern coal seams. On this basis it appears that use of physical coal cleaning for pyrites removal may have a more profound effect on FGD economics than merely the cost saving calculated from the resulting decrease in SO_2-removal load. A possibly significant additional justification for supplemental physical cleaning of coal is the savings in capital charges and manpower and material cost associated with reduced maintenance and unavailability. Moreover, as is not commonly recognized in the conventional coal-energy field, efficient physical cleaning of coal is highly advantageous, separate and apart from

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the benefits of reductions achieved in ash and sulfur content, because of the day-to-day uniformity in the quality and fuel characteristics of washed coal and the resulting improvement in operation and reliability of the entire power plant.

(4) Access to Waste Disposal Space

As in the case of 13% of FO-FGD capacity purchased in U.S.A. to date, feasibility of some candidate retrofit sites may depend vitally on the use of a commercially available and economically feasible by-product yielding FGD process. The most notable candidate process is FO-FGD forming salable gypsum by-product. A number of factors hindering selection and economical use of by-product gypsum FGD together with potential solutions tied to overseas experience and advancements with this common worldwide by-product process are outlined as follows:

a. Achievement of Gypsum Quality Criteria in Medium/High Chloride Coal Service--It is necessary to predicate the design of one or more of the U.S. by-product FGD gypsum systems on using washed-cake repulping following by repeat filtration/wash. This cumbersome arrangement may be avoidable in future applications through use of a one-of-a-kind West German FO-FGD technology commercially applied at West German electric power company, NWK, which yields unusually large (300 micron) gypsum crystals that are washed in conjunction with a single filter-step at extremely high (90-95%) cake washing efficiency, to yield a wallboard quality product.

b. Moisture in By-Product--8to 12% weight surface moisture in the typical by-product detracts significantly form the sales value of the material. A cost-effective West German method (using gas cyclones) of using a small proportion of the 300° F boiler gas to evaporate and remove the bulk of the free moisture in FGD gypsum cake has been successfully piloted by West German electric power company, STEAG, and systems of this type are now commercially available.

c. Need for Agglomeration of By-Product--In many applications acceptability and sale of the gypsum will be contingent upon its being agglomerated before shipment from the power plant. Based on an evaluation by a U.S. power company that has purchased a by-product FGD system, a highly effective and economical West German agglomeration-by-extrusion process currently being applied for this same purpose by West German electric power company, Isar-Amperwerke, appears to be an optimal technology to meet this need in U.S.A.

d. Expansion of Market for High-Value Gypsum Products--With major increase in longwall-type underground coal mining in eastern U.S.A. a large market is being created for a normally costly gypsum species, the alpha crystal form of calcium sulfate hemihydrate. Retrofit candidates who elect to apply a by-product mode can derive a major cost benefit by serving this market. Based on advancements in West Germany in ways to prevent subsidance caused by longwall mining, it may be anticipated,worldwide, that increasingly large tonnage quantities of alpha hemihydrate will be in demand for use in admixture with coal ash to implement underground structural support techniques for subsidance control. An FGD-related horizontal-belt-filter

mounted by-product alpha yielding process that substitutes for a normal by-product gypsum dewatering and cake-washing filter has been developed[19] and is now undergoing small scale field testing at a West German FGD (STEAG) installation under the sponsorship of the West German government.

e. Impact of Gypsum Depletion Allowance of IRS--The Internal Revenue Code, Section 613B, includes gypsum[20] among a number of other mined mineral resources for which major income tax benefits are granted to mining companies to compensate them for depletion of mine holdings. This serves as a disincentive to the use of by-product gypsum since the allowance in effect, drastically and artificially reduces the cost of the mined natural gypsum. Experience in Japan and West Germany, where markets for by-product gypsum are brisk, suggest that promotion of a free U.S. market in gypsum could contribute substantially fo increasing both the demand and the market price for by-product gypsum to the betterment of the ongoing and pending emission control programs in U.S.A.

(5) Upgrading of Existing Electrostatic Precipitators As Necessary

FGD design strategy may call for improving the performance capability of existing fly ash collectors. There will, in any instance, be many practical means of upgrading the equipment to near-new collection efficiency. Comparatively new but extensive overseas design capabilities, particularly in Japan, for wide-spaced collecting electrodes, may be applicable in some cases since a major increase in potential particulate collection efficiency is possible for a given housing size.

(6) Receiving Streams for Liquid Effluent

With no acceptable means of outboarding a necessary blowdown of saline FGD liquor purge, West German emphasis on direct-contact flue-gas reheat recovery technologies may offer or lead to a design means of utilizing a proportion of 300° F flue gas in a spray dryer arrangement to convert discarded treated-liquor or brine to a salt cake, principally calcium chloride, that would be utilized as a by-product or discarded in a small-scale secure landfill operation.

(7) Protection of Existing Stacks

Wet-scrubber outlet-gas reheat sufficient to protect the liner of an existing tall stack should be incorporated in the most cost-efficient manner. Where remaining plant lifetime is sufficient to justify it, this may call for use of commercial overseas technology for regenerative flue gas reheat using an adaptation of the rotary air preheater. Many have been installed in Japan new coal-fired units. The largest in the world, of 350 MW equivalent capacity has operated successfully for over a year[21] providing in excess of 100° F reheat at an FGD installation of West German power Company, NWK.

Technologically advanced dynamic mist eliminator designs of German origin, now exploited only sparingly in USA, offer a valuable adjunct to reheat use

B. DRY SCRUBBING

(1) SO_2 Removal

Based on early FGD system experience with high-sulfur coal service principally by 1 or more Danish firms serving the U.S. FGD market, significant retrofit application of lime spray drying appears possible in site-specific applications in which each of the following holds:

- Plot space availability at the back-end of the boiler will accommodate both the spray dryer facility and the required dry particulate collector downstream of it.
- Coal sulfur level is within the practical upper limit, if any, of the range of values at which the dry scrubbing technology has adequate performance capability and system reliability. Based on sustained runs on high-sulfur Illinois coal in an industrial boiler application the process is capable of high removal efficiency at coal sulfur levels as high as 4.5%. Moreover because of the uniqueness of spray dryers (when contrasted with wet scrubbers) as gas/liquid contactors for removing SO_2 from boiler flue gas, system reliability should be no more impaired by process-related factors in high-sulfur applications than in the more common low-sulfur applications; dry scrubbing avoids water saturation of the flue gas and related sulfurous acid and hydrochloric acid corrosion effects and, moreover, because of uniquely high SO_3 removal capability, consistently reduces flue-gas-outlet sulfuric acid dewpoint temperature to approximately the same uniform level[22] irrespective of coal-sulfur level and SO_2 and SO_3 concentrations at the system gas inlet.
- Solid waste management by throwaway disposal is feasible.

The justification and advantage of dry scrubbing will be increased when any or all of the following site conditions prevail:

- A short remaining plant lifetime seriously detracts from a higher-capital cost solution such as wet scrubbing.
- Existing fly ash collection equipment has poor performance or reliability and/or its removal from the plant would serve the purpose of providing space to accommodate FGD.
- Absence of natural water bodies adequate to receive and assimilate treated wet-scrubber FGD liquor-blowdown seriously detracts from the economic evaluation of open-loop FGD schemes such as FO-FGD. This will be particularly true in high chloride coal service, which generally results in increased operating/maintenance problems in wet scrubbers leading to need for increased blowdown, and in which case dry scrubbers are reported to have improved performance[23].
- The amount of "free" flue gas reheat afforded by dry scrubbing system operation significantly reduces the cost of flue gas reheating otherwise required to protect the existing stack.

(2) NO_x Removal [24]

One of these firms report that use of sodium hydroxide (10 weight % of lime feed) additive for high SO_2-collection-efficiency systems operated at

elevated lime-spray-dryer outlet gas temperature (approximately 200° F), yielded at least 50% NO_x removal in a high-sulfur industrial boiler demonstration test. It appears that this improvement is specifically applicable to high-sulfur applications.

C. LIMB

Limestone injection into boilers, an FGD method that was field assessed and abandoned in U.S. in the 1960s, has gained new interest in U.S. as a result of success achieved in West Germany in recent years with lime injection and multi-stage burners (LIMB). It appears that ongoing test work in Germany will provide early guidance as to the extent of applicability of this low-capital cost method in U.S.A.

(1) Application to Lignite-Fired Utility Boilers

West German electric power company, RWE has reported to U.S. EPA attractive results in SO_2 and NO_x reduction using a method of lime injection and multi-stage burners it has developed and demonstrated on a German lignite, i.e. brown coal, fired boiler. In view of the extremely high chemically-bound moisture content of the German brown coal, substantially higher than even U.S. lignite, its combustion temperature characteristics are seen to favor easy application of the LIMB principle. Accordingly, for purposes of assessing applicability of this technique to the U.S. bituminous coal-fired units, the RWE work is of value primarily in establishment of a baseline comparison of other ongoing tests on boilers firing more relevant fuel types.

(2) Application to German Hard Coal [25]

West Berlin utility company investigators report initial small scale (2.3 MW) tests on German hard (stone) coal, similar to bituminous. Activated hydrated lime was used with 50% utilization efficiency. Instead of injecting alkali with fuel as in earlier brown coal tests, the lime was

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... (continued from page V-3)...
so as to adequately limit fouling of diverse reheat devices that may be used; or merely to prevent excessive carryover from wet scrubbers of scrubbing slurry droplets that can aggravate solid particulate emissions and/or downstream corrosion on low-pH inner duct walls and stack-liner walls. All such German designs, as well as U.S. facsimiles also available here, provide unique liquid drainage columns incorporated in aerodynamically designed eliminator vanes or passages and permit the use of elevated design gas superficial-velocity in this system component so as to more effectively trap out critically important fine droplets without reentraining the liquid that is routinely collected on the internal eliminator surfaces.

injected via peripheral jets arranged around the burners. Additional testing was done on a 700 MW unit, converting two of the 24 burners for trial LIMB operation. The results justified converting all 24 burners to LIMB. This adaptation will be completed in 1983 and full scale testing will be performed in 1984. The Berlin workers cite the economic advantages of this concept for small power plants with low capacity factors. They indicate that the control method is expected to be applicable in Germany for all old boilers either singly or in combination with wet scrubbing

D. NO_x CONTROL BY COMBUSTION MODIFICATION

Dr. Ando [26] reviews the very extensive work in Japan pointing to and emphasizing low-NO_x burners of Japanese development, which, installed in conjunction with other combustion modifications, typically limit NO_x emissions from large coal-fired utility boilers to approximately 200 ppm. Possible advantageous retrofit of any selected Japanese burner technology depends on the configuration of existing boilers and the extent of development of equivalent retrofit techniques by the U.S. manufacturer of the individual boiler. All U.S. domestically available retrofit combustion-modifications methods for coal-fired boilers have strong economic incentives [27] in any scenario in which optional NO_x control is credited against an SO_2 removal objective.

E. MISCELLANEOUS

Numerous experimental and developmental NO_x-only and SO_2/NO_x removal processes, wet and dry, not commercially applied and not inter-related with commercially available and applicable SO_2 removal processes are excluded from this review. They are not within the scope of this engineering oriented overview/assessment of available overseas technologies useful to retrofit applications.

VI. POTENTIAL GOVERNMENTAL ACTION STEPS TO OPTIMIZE TECHNOLOGY SELECTION AND APPLICATION

Significant advancements in the body of U.S. technology and know-how in design of systems for SO_2 and NO_x control may be expected to come about through foreign technology transfer merely by the use of all available forums for publicizing all good work accomplished abroad. This effort would be greatly strengthened by your full-time use and deployment of cognizant professional engineers at the two major overseas "frontiers", Japan and West Germany, to assure fullest awareness and comprehension of the many relevant technical-development and commercial-application milestones that are frequently being reached there through the truly impressive technological mentality and engineering skills in those regions of the world. Opportunities for effective governmental action in an even more concerted and focused manner seem to me, from the vantage point offered herein, to be self-evident.

A. SUPPORT OF ADVANCEMENT IN TECHNOLOGY USED

Our federal-government supported SO_2 and NO_x control R & D has been a continuing and concerted effort by many skilled and dedicated people in EPA, DOE and other agencies. But its effectiveness in advancing technology utilization in our electric utility industry could, I believe, be very substantially advanced by an increased emphasis on programs concerned, apart from true technical development, with identifying and, more importantly, harnessing already available technologies; particularly those from abroad, that can/will permit us to leapfrog much paralleling do-it-yourself development effort by our own workers. Our active participation in as well as continuing scrutiny of overseas technical and new development in the field, some of which has been identified herein, appears to me to offer large rewards. New 1983 R & D reports on Japan by EPA, referenced as (2) and (26) in Section VII, give impressive testimony to what is already being accomplished through continuing consultation by Dr. Ando of Tokyo's Chuo University in cataloging and describing that country's growing technology base. In view of the great strides that now also have been made in West Germany, a comparable zeal should be directed to studying, evaluating, and continually updating ourselves on the extent of that country's accomplishments.

B. SUPPLEMENTAL LEGISLATIVE ACTIONS

Specific examples of new federal legislation that would significantly improve prospects for cost-effectiveness in potential capital programs and maximize protection of the environment at plant retrofit sites are offered here.

(1) Licensing of Electric-Utility Field Personnel

Congress has an important opportunity to help upgrade the manner of operation and the on-line reliability of all future (and present) gas cleaning systems in the regulated electric utility industry through legislative or administrative steps that would provide for improved training/

recognition as well as licensing of pollution-control-system field personnel. Members of a recent inter-agency task force to Japan investigating some aspects of air pollution control technology utilization there have brought back valuable details of a truly massive personnel training and self-schooling program spanning more than ten years that appears to account in great measure for the well-publicized and outstanding record of on-line reliability of Japanese FGD installations. Japanese federal law initiated in 1971 requires manning of operation of commercial air pollution control installations by an adequate number of nationally certified college-bred or practical engineers. Some of the details are as follows:

- A national examination system periodically offers comprehensive written exams in the individual areas of air pollution control, water pollution control and others.
- A 700 page national handbook, revised every four years, is used as the "bible" and serves as a training aid.
- On the average, each time examinations are offered, 30% pass the test and receive certification.
- 350,000 certificates licenses have been issued, 40% in air, 40% in water, 20% in other.
- The certification program was organized and put in place by Dr. Masuda of the University of Tokyo, world reknowned electrostatic precipitator expert.

This program and the professional/societal rewards and recognition that it extends to licensees is clearly a part of the Japanese "can do" approach to FGD success. This writer and others who have repeatedly studied Japanese operations in the field can attest to the implantation in FGD operators of right mentality/attitude, competence, diligence, reliability, dedication to environmental objectives, and work-ethique.

(2) Abridging of IRS-Code Depletion Allowance Applicable to Minerals Available as FGD By-Products

U.S. Internal Revenue Service Code Section 613B provides for a substantial tax benefit (allowance) tied to an owner's depletion of mineral resources. In recognition of the strategic value of natural gypsum deposits in North America, this mineral is included among those given this treatment. The result is that the net cost to the mine owner in his use of a naturally plentiful natural raw material to manufacture finished products is substantially reduced in an artificial manner. This significantly detracts from the potential selling price of by-product gypsum from a power plant source and is a major deterrant to the extensive use of FO-FGD technology for disposal-by-sale of a gypsum by-product yield enjoyed throughout the principal overseas countries to which this overview is directed. No spokesman from the pollution control system applications sector would want solutions to FGD system optimization and solid waste management problems to culminate in a financial hardship for the North American gypsum industry, already depressed by current reduced demand/production. Congress can authorize that IRS or another agency to find a way to eliminate the gypsum depletion allowance without a net financial tax debit to those now sharing in this tax break; giving added incentive points of consumption in the huge captive gypsum market in U.S.A. to negotiate for purchase of FGD by-product gypsum on a free-market basis.

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Biographical Sketch of

WILLIAM ELLISON

William Ellison PE has served since 1981 as Director of independent consultant, Ellison Consultants, Monrovia, Maryland, serving domestic and overseas clients. His field of specialization for the past seventeen years has been stack gas cleaning and management of resulting liquid and solid wastes. He has thirty years of experience with engineering-construction companies, air pollution control system suppliers and consulting engineering firms in technical development, design and operation of process and electric utility plant facilities.

Ellison is a professional engineer licensed in the State of New York and Pennsylvania. On a number of occasions he has served as an expert witness before governmental regulatory agencies including air pollution control boards and since 1970 has made frequent presentations in technical journals and conferences addressing technology for environmental pollution control in operation of coal-fired boilers.

His educational background includes the degree of Mechanical Engineer and of Master of Science in Heat Power, (Stevens Institute of Technology), as well as Master of Engineering, majoring in Environmental Pollution Control, (City University of New York.)

Mr. FUQUA. Thank you, sir.

Dr. El-Ashry and Dr. Yeager, we have been spending—well, this year we authorized some $2 million for the Grimethorpe pressurized, fluidized bed in England. Has there been anything that TVA or EPRI has received from that that's worthy? Should we continue with that or——

Dr. EL-ASHRY. I'll let Mr. Yeager answer that.

Dr. YEAGER. Yes, sir, I think as I testified to the committee in previous visits, we view that as one of the most important contributions of the Federal investment in the energy R&D area.

The Grimethorpe program is the key to our ability to understand how to design and operate pressurized, fluidized bed powerplants. Their value in this context is the opportunity to build modular power units that can be installed rapidly in the field, operating on coal to very high levels of environmental control, and at quite low cost by virtue of building them very quickly.

So we view, from the utility industry standpoint, a very important aspect of that technology, and it's one that we are hopeful we can pursue. We have in our plan the opportunity to build with manufacturers in this country a demonstration of that unit. That demonstration, however, is totally dependent on the information coming from Grimethorpe, and we feel that it must run for at least another 1 to 2 years to resolve the issues.

Mr. FUQUA. Dr. High, maybe you can—I know that's a pressurized bed, where you're talking about an atmospheric bed, but is there any technology that—or demonstration of technology—that would be helpful to TVA and your plant at Paducah?

Dr. HIGH. Yes, sir, I think so. The fundamental research that went into all the fluid bed facilities, including Grimethorpe, has been essential in the development of fluid beds, including the atmospheric. There are some unique problems in the pressurized version that are not present in the atmospheric, as you might expect.

We also view the Grimethorpe facility as an essential element in the development of pressurized fluid beds.

Mr. FUQUA. Dr. El-Ashry, how much nuclear power—what percentage of TVA output is produced by nuclear power?

Dr. EL-ASHRY. It's about 30 percent, sir.

Mr. FUQUA. Thirty percent?

Dr. EL-ASHRY. Thirty percent.

Mr. FUQUA. What would be the effect if this 30 percent were produced by coal-fired plants for the SO_2 emissions?

Dr. EL-ASHRY. I believe it would be about 300,000 tons. I just take one-third of our output at the present time.

Mr. FUQUA. And you plan to add other nuclear plants to your generation capacity?

Dr. EL-ASHRY. We have four units under construction nearing completion at the present time.

Mr. FUQUA. And what will that raise the percentage of nuclear power produced compared to 30 percent today?

Dr. EL-ASHRY. Probably about 45 percent.

Mr. FUQUA. So it would be a significant reduction in the SO_2 when those come on the line?

Dr. EL-ASHRY. Yes, sir, and that's exactly what figures out in the 8-million-ton reduction national goal that would not cost the TVA ratepayers anything.

Mr. FUQUA. Dr. Yeager, you gave an illustration of the amount of residue from a scrubber of a 1,000-megawatt plant, I believe. What would be the amount of spoil from a fluidized bed, the gypsum or the limestone or the dolomite, whatever source was used, for a similar type plant?

Dr. YEAGER. This—the byproduct from the fluid bed, the fundamental difference is it's totally dry, which has a major impact on the volume of it. So it would be the basic fly ash plus the amount of limestone associated with it, and we are now in the range of about two units of calcium, if you will, or limestone to unit of sulfur. So we would be about a 50-percent increase in the byproduct loading relative to fly ash alone, but about one-half that which would come from a scrubbed powerplant.

Mr. FUQUA. In the National Academy of Sciences recent report about acid deposition, they were talking about the nonlinear relationship between long-term average emissions and deposition. There's been some scientists from some of the DOE and national labs that have disagreed with both the methods used to reach this conclusion and also with the conclusion itself. What is your opinion? Do you agree with the conclusions of the Academy panel or do you think some of their information was faulty?

Dr. YEAGER. Well, I would prefer, sir, to perhaps pass that on to the experts in our group with that. I'm really on the control sides of things.

I think that certainly all of the evidence I have seen in this area would indicate that it is not just the long term but the near term, the near close-in sources that certainly have to be considered very strongly in this area, and I think that there is a great deal that isn't known about the relationship between sources and deposition, and that, I think, is the key to the uncertainty in the whole science of this area, and one that needs a major approach if we're going to understand what an investment at one source, in terms of control, is going to mean to a reduction in deposition at another point.

Mr. FUQUA. What—it is my understanding that EPRI does have an interest in the pressurized, fluidized bed at Woodridge, N.J. What—what role is EPRI playing in that?

Dr. YEAGER. Well, we have been very, very interested in that particular unit because, again, we see it as another element that would help provide the technical basis for the industry to apply this technology.

That facility is one in which we have collaborated in the technical aspect of it. We have not participated financially in the program, and at this point would not plan to until it has shown its ability to operate successfully. It is still in construction under DOE auspices at this point.

Mr. FUQUA. Mr. Ellison, you've noted that the ambient air quality in Japan for SO_2 and nitrogen oxides are substantially tighter than the U.S. criteria. What comparable figures do we have for that?

Mr. ELLISON. In the text of my testimony the figures from Japan are given, if I can quickly find them, Mr. Chairman.

That would be under section 2, I believe.

In Japan the ambient air quality standards expressed in terms of yearly average, and this is quoting from Dr. Andro's work, 0.02 ppm for SO_2, approximately 0.02, and for NO_x somewhere in the range of 0.02 to 0.03, as he expresses it. This is not quite half, but perhaps maybe 75—60 percent of the allowable numbers here. Someone may be able to correct me on that, but substantially lower. And Dr. Andro has been quick to emphasize this to explain in part the zeal with which Japan has gone about retrofits and the major program that they've achieved.

Mr. FUQUA. You also mentioned the contrast between Japanese and European plants and the Eastern U.S. electric generating units that use coal fuel that had a high sulfur content of between 3 and 5 percent, and that, as a result, the flue gas desulfurization reliability was only 81 to 82 percent. What is the average reliability of the Japanese and European similar-type facilities, the flue gas desulfurization?

Mr. ELLISON. Japan—and, again, we're quoting Dr. Andro's voluminous details and records—97, 98 percent in Japan. In good measure, on low-sulfur applications, where wet scrubbers—the Achilles heel of wet scrubbers in terms of the acidness syndrome are not nearly so noticeable or troublesome.

In the case of Germany, there are so few units in the new program already running that we don't have too many numbers, but it's—the numbers I do have on one or two indicate that we're up in the high 90's there as well, again on comparatively low-sulfur fuels where wet scrubbers have a far better place in our list of tools.

Mr. FUQUA. Mr. Gregg?

Mr. GREGG. Thank you, Mr. Chairman.

Mr. High, I notice that you felt that TVA could take its share of the 12-million-ton national reduction, and I assume you're referring to the 31-State region, with about a 4- to 6-percent increase in energy costs in the TVA region, by shifting to other generating methods and conservation efforts and having the ability to trade within States.

Shifting to other generating methods, does that mean going to nuclear basically?

Dr. EL-ASHRY. Yes, that's taking credit for the nuclear capacity that could replace coal-fired——

Mr. GREGG. The point you were discussing with the chairman earlier?

Dr. EL-ASHRY. Yes.

Mr. GREGG. And conservation efforts, you're just projecting a lower usage over that timeframe or a less growth than you would have—you normally project?

Dr. EL-ASHRY. Actually, a higher level of conservation than we have had, but mainly the projection from the effort that has been going on for the last 6 years.

Mr. GREGG. So you don't think you have to go to any major control technology in order to accomplish some sort of a reduction?

Dr. EL-ASHRY. If our loads remain essentially about where they are now, at the lower to medium forecast, as we say.

Dr. HIGH. Can I add a statement to that?

Mr. GREGG. Yes.

Dr. HIGH. We plan in the conservation area, not just conservation from price elasticity but from conservation efforts that we've done, to have an equivalent of about 1,400 megawatts in the early 1990's.

Mr. GREGG. Thank you.

Mr. Yeager, you were talking about fluidized beds with the chairman. What's the wear on a boiler from a fluidized bed? Does the boiler have a different life span if it's using a fluidized bed than one that would not—which would, say, use flue scrubbers?

Dr. YEAGER. Well, of course, that's one of the key points that the development program that we have underway today is designed to find out. The experience that we've had to date, both in small pilot units as well as the unit at TVA, would not show that erosion rates or other effects from the operation of the fluid bed would change relative to a conventional plant. That's largely a matter of knowing what materials to select and how to select them. So the boiler itself seems not to be affected reliability-wise.

Dr. High, would you want to add to that?

Dr. HIGH. No. I think that's correct. We haven't seen any evidence at the pilot plant, with the 3,200 hours or so, that that erosion in the boiler itself is going to be a problem. There have been units built in the industrial sector that have shown erosion, but we haven't experienced that. I think it's a matter of material selection and how you put the material in the boiler.

Mr. GREGG. Do you see fluidized beds as a viable option—yes or no, I guess? I would like that sort of response.

Dr. HIGH. Yes.

Dr. YEAGER. Yes.

Dr. HIGH. I don't know what a viable option to, but, yes, it's a viable option.

Mr. GREGG. Well, to, say, scrubbers?

Dr. HIGH. Most certainly, yes. You have to put that in perspective, however. In the near term scrubber technology is, for the large part, commercial, even though there's reliability and availability problems. The fluid bed for the industrial sector will not be available for—until the mid-nineties. But from that point forward, we certainly think the fluid bed is a coal-fired option.

Mr. GREGG. Do you see LIMB as ever being—do you see LIMB as falling in that scenario, also?

Dr. YEAGER. Well, I think that LIMB, we would view—or the industry would view—as a potential, and again speculative at this point, technology for application to the existing boilers that are out there, those existing plants, particularly ones with very limited future lifetimes where the installation of major capital modifications is just not going to be justified. So in order to keep that capacity available, it may have some attractiveness. I underscore, though, that it is, in our judgment, not available to the whole population of boilers out there. It will be some fraction of that which we're still determining.

Mr. GREGG. How far away are we from using LIMB for those boilers which have a limited life?

Dr. YEAGER. Well, our plans would carry us through—if each of the preceding steps were successful—large-scale—and I'm talking by that several hundred megawatt scale boiler—demonstrations by

late this decade, so that by 1990 or in that time, let's use that as a good figure, the technology would be well defined for commercial application. I do not think it would be available on a confident basis before 1990 for the high-sulfur boilers that we're faced with here.

I'd like to underscore, if I could, just one point there, because there is a lot of attention to what's going on in Germany, and certainly we're very close to and working with the Germans, but let me underscore that high-sulfur coal to them is less than 1 percent. The issues of this technology on a low-sulfur coal versus a high-sulfur coal, both in terms of the quantities and the ash characteristics, are substantial.

Mr. GREGG. But you do feel that it will be a viable technology by 1990?

Dr. YEAGER. Well, we certainly hope so because it's a good deal more cost-effective in many potentials, if it works, than putting a scrubber on. So there's a lot of self-interest in trying to make it work.

Mr. GREGG. To the extent that new powerplants are developed, do you expect them to be the major central powerplants that we've seen historically or are they going to be smaller plants spread out over larger regions?

Dr. YEAGER. Well, you'd probably get a lot of different answers on that question. It depends a bit on the scenario that you want to paint.

I think that clearly the industry's interest right now, because of the difficulty of acquiring capital and the limitations on growth of electricity, that there is a good deal more interest than there was, say, 5 or 10 years ago in being able to build smaller, modular power units that can match the load growth, can be built quickly to cut down the cost of work in progress, and, hence, be an overall much more economical approach. And that, of course, is in turn the emphasis the industry is putting on the pressurized, fluidized bed technology, and also the atmospheric, both of which lend themselves to a greater degree to that option than the conventional technology we use today for coal.

Mr. GREGG. That was going to be my next question. So you see those as the two technologies that would apply to your smaller plants?

Dr. YEAGER. We think that they're the leading ones. There's certainly other options. In some situations coal conversion may have a role in that area. As we've gone through the comparative economics, in our judgment, the—particularly, the modular, what we call, turbo-charged boiler appears to have the greatest potential for that kind of economy—reverse economy of scale, if you will.

Mr. GREGG. Do you see any nuclear plants being built in our lifetime?

Dr. YEAGER. That, again—I certainly hope so, in my lifetime, anyway. I think we're going to be in deep trouble energywise in this country if we do not find a way to resurrect the nuclear initiative. I think that for the remainder of this century I see little evidence, other than the plants that are in the pipeline, that you're going to resurrect it, but I think that it behooves the Nation to do the research and development that are necessary to come up with

a nuclear cycle that is compatible with all of the public concerns that have been registered. I don't think that's an impossibility. I think we need to make the effort.

Mr. GREGG. But in the timeframe that we're trying to address acid precipitation, you don't see the plants——

Dr. YEAGER. I think that if—I see no evidence of that, unfortunately, being the case. I think that many, many units are being canceled, have been canceled; many more will be. The roadblocks in the pace of putting those plants in has just become very large, although, as it has been pointed out by TVA, a key element of their reduction strategy, if you will, is the nuclear aspect of it, and that that is an important tradeoff in these areas.

Dr. EL-ASHRY. Let me just correct this: That was not—the strategy of building nuclear powerplants was not for the purpose of reducing SO_2 emission.

Dr. YEAGER. It is a valuable side benefit, though, I think.

Mr. GREGG. Thank you.

Mr. FUQUA. Mr. Lewis?

Mr. LEWIS. Dr. El-Ashry, recently there have been some strong statements that work must begin immediately to curtail the damage from acid rain without waiting for research to be completed. And if such a position became a directive from the administration, what actions would TVA take to participate and in what kinds of activities?

Dr. EL-ASHRY. You mean actions to control SO_2?

Mr. LEWIS. Yes.

Dr. EL-ASHRY. Basically, as I said in my testimony, depending on the goal, we are willing to take our fair share, and we propose two major points really, and that's the flexibility, and flexibility in applying whatever goal that is there, and then the second point is allowing utilities, particularly large utilities in multi-States, to have also the flexibility to take their fair share wherever they may choose.

Mr. LEWIS. I see. Well, with—in your prepared statement, which acknowledges the lack of acid rain definition, why do you advocate further reductions in sulfur and nitrogen oxide emissions?

Dr. EL-ASHRY. Well, I don't think, you know, we say that there's no acid rain definition. We say that——

Mr. LEWIS. Well, there's a lack of definition. I think there is a definition, but there is a lack of the expressed definition that——

Dr. EL-ASHRY. What is lacking is a direct link between the causes and the effects. And what we say is that it might—we may never get to that point of finding a direct link, because of the complexity of the natural system that acid rain is only part of. But the circumstantial evidence seems to be overwhelming, and we believe that we may be risking some irreversible damage as a result of all the things that we're seeing right now not only in the United States, but in Europe as well.

Mr. LEWIS. I see. I guess I'm going to also ask you this question, along with Mr. Ellison: I'm having a problem personally trying to look at the acid rain problem overall in the United States, when we're talking about losing lakes and killing off the fish, and from the time of the industrial revolution we've had tremendous stack emissions in the United States as well as other countries. And I'm

wondering if there's been any research done or you have made any studies into blocking out, in segments or time frames, the amount of atmospheric contamination due to acid rain and whether it was greater 50 years ago, 60 years ago, percentagewise than it is today, and if so—if not, what would be the chances of overcontamination, we'd say, within the next 20 to 30 years, because we're still trying to determine the definition of acid rain to a great extent?

Dr. EL-ASHRY. I do not know of anyone who has separated or can separate these points as clear as they can be done. However, we look at trends, and the trends have been toward increased acidity of the rain as well as the aerial extent for that acid rain, and particularly toward the Southeast, and that's what concerns us the most.

In terms of projecting it into the future, I think it's a matter of believing in these precursors for acid rain, and as you would increase the emissions, you would increase the acidity of the rain and the aerial distribution of that.

If, as the National Academy of Science report has shown, if you would cut these emissions, then you would get similar reduction in the acidity of the rain that would fall as a result of this cut.

Mr. LEWIS. Would you care to comment, Mr. Ellison?

Mr. ELLISON. A well-informed geologist would, I think, be far more competent to discuss this than I, but I—from my own reading on the subject, it seems to me that the geotechnical, geological people who've talked about the acid rain impact areas seem to be coming very close to telling us that during the course of the industrial revolution we've managed to, through acid rain percolation, we've managed to devour a comparatively thin layer of what the geologists call karst, k-a-r-s-t, better known as limestone, and have now in many areas which we call sensitive areas, that were either sensitive when we started or have become sensitized, we have large areas now that have done the job—have depleted their karst layer and are no longer able, like a tooth in your mouth with the enamel worn out, is no longer able to take the gaff of daily existence.

That's my—that's the picture I get. I'm baffled as to why our geologists have come so close to telling us what I just said without saying it.

Mr. LEWIS. Dr. Yeager, would you care to comment on that? Has your institute done any work in this area?

Dr. YEAGER. Yes. Certainly we have examined the properties and, if you will, the buffering capacity of a number of the regions that are affected and are continuing to do so. And I would say that the results—and, again, I am not necessarily the best one to interpret it, but I'll give you my view on it—is that they—they're a very mixed bag of results, and they're largely a mixed bag, again, because of the local geology, but also the local biology.

We must remember that there are a lot of sources for acidity into the ecosystem, and that the activities of man are certainly not minuscule in that regard, but by no means are they the total picture.

And in many of the lakes that we have found, where one has gone through the sediment in the bottom of the lakes and made examinations of what has been the history in terms of the acidic action found through those sediments, and one has found that that

history has been very long and it has been related as much to biological activity as to anything else, the degradation of the biota in those particular areas.

So one has to—that's an area where there is a great deal of work to be done, and it is one of the elements that we have recommended to EPA and DOE receive a good deal more effort, so that we understand what the results will be of any kind of a control effort we take.

We have to also remember that the sources, the amount of acid we put into the atmosphere—when you change—when you—if you even have the amount of material going into the atmosphere, the effect on acidity as we would measure it by pH is almost minuscule. It's measured in tenths. The issue is really the deposition of sulfates and so forth, and there I think there is a much stronger case to be made for the reduction of emissions, although what you reduce and where the effect will occur, we don't know at this point, and I don't think any of the scientists—if there's a common thread, it's that they don't understand that relationship at this point.

Mr. LEWIS. Thank you.

Thank you, Mr. Chairman.

Mr. FUQUA. Thank you, Mr. Lewis.

And we want to thank you for being here, and we appreciate your testimony this morning. Thank you.

Our next witness will be Dr. Frank Princiotta, Director of Industrial Environment Research, Office of Research and Development, EPA, and Jan Mares, Assistant Secretary of Fossil Energy of the U.S. Department of Energy.

Dr. Princiotta, we'd be pleased to hear from you this morning.

STATEMENT OF DR. FRANK T. PRINCIOTTA, DIRECTOR, INDUSTRIAL ENVIRONMENTAL RESEARCH LABORATORY, OFFICE OF RESEARCH AND DEVELOPMENT, U.S. ENVIRONMENTAL PROTECTION AGENCY

Dr. PRINCIOTTA. Thank you very much.

Good morning, Mr. Chairman and members of the subcommittees.

My name is Frank Princiotta. I am the Director of the Industrial Environmental Research Laboratory of EPA in North Carolina.

I appreciate the opportunity to meet with you and discuss several forms of existing and developing technology designed to control sulfur oxides and nitrogen oxides. Based on our current state of knowledge, these pollutants appear to be the major precursors to acid deposition.

Emissions from fossil fuel powerplants have been the focus of attention for possible approaches to controlling the release of acid precursors, because nationally these sources produce a major share of the sulfur dioxide and a sizable portion of nitrogen oxide emissions. In addition, other activities, such as smelting, industrial combustion, transportation and space heating, when viewed together, can contribute significantly to the atmospheric loading of both sulfur and nitrogen acid precursors.

My comments will focus primarily on coal-fired utility boilers because our knowledge regarding the control of these sources is more

complete. I will also discuss the technologies capable of being retrofitted to existing boilers primarily, since control of such sources are likely to be an important component of any future acid rain precursor control program.

I have included in my testimony two tables that summarize most of my remarks. Table 1, for example, summarizes the major manmade acid deposition precursor pollutants, the major industrial sources responsible for those pollutants, applicable controls, and a very brief statement explaining their operation.

Table 2 focuses on coal-fired powerplants, which, as I indicated, are the major emitters of both sulfur oxides and nitrogen oxides. It's among the stationary sources. This table summarizes performance, commercial status, estimated costs, and applicability for available and emerging control approaches. Again, the emphasis here is retrofit or application to existing sources of these precursor pollutants.

My following discussion will elaborate on the state of the art and the Federal research program relating to these two major acid deposition precursor pollutants.

Moving first to sulfur oxide control, the main approach is through reducing emissions of sulfur oxides from coal combustion other than switching to low-sulfur fuels are removing the sulfur from the coal, capturing the sulfur oxides during the combustion process, or removing or scrubbing the sulfur oxides from the flue gases resulting from the combustion.

Of these methods, flue gas scrubbing, we've heard quite a bit about that already, also called flue gas desulfurization, is really the only technology that is used commercially on a wide scale at electric utility plants. To date, more than 100 such installations, scrubbing about 44,000 megawatts of capacity, are installed. If we add planned capacity, we expect over 200 utility FGD units to control over 100,000 megawatts of capacity in this country by the turn of the century.

These scrubber processes can be classified as those that produce a waste product—we've heard quite a bit about the disposal of that product so far—versus those that produce a saleable product, such as sulfur or sulfuric acid. Almost all the current and planned FGD installations in the United States, at least, are the waste-producing type, because generally these are the most simple and least expensive processes.

The most common processes that produce a waste product are those scrubbers employing lime or limestone as the alkali reagent. These reagents react with the acidic SO_2, yielding insoluble calcium salt wastes, generally insoluble materials. The processes that produce saleable products are more complicated and normally more expensive. The cost of the current flue gas desulfurization is quite high in terms of total dollars and represents the most expensive air pollution control device for coal combustors. Typically, such costs for retrofits might be in the order of $200 to $300 per installed kilowatt. This compares to roughly a thousand dollars a kilowatt for a new coal-fired powerplant.

Since the early 1970's, EPA's research program has played a crucial role in upgrading the performance and reliability of the lime

and limestone scrubbing technology. We've worked in concert with TVA and industrial sponsors, I should point out.

EPA has recently demonstrated the ability of organic acid additives, adipic acid being the most important, to further improve performance and reliability over state-of-the-art scrubbers. At EPA research efforts continue, albeit at a relatively low level, to seek lower cost FGD processes with particularly emphasis on spray dryer flue gas desulfurization systems, which offer the potential for significantly lower capital costs especially than corresponding wet scrubbers.

Turning to coal cleaning, physical coal cleaning represents an alternative and/or supplemental sulfur oxide control approach. Coal is physically cleaned, a process sometimes called washing, by crushing and utilizing separation equipment to remove some of the mineral impurities, including inorganic sulfur. Conventional coal-cleaning processes can reduce the potential sulfur emission from the combustion of coal typically by only 10 to 30 percent, depending very strongly on the specific coal involved.

Approximately 30 percent of all utility coals are cleaned. Current practices reduce the potential annual SO_2 emissions from coal production by about 1.8 million tons. Conventional coal-cleaning processes increase the cost of producing electricity somewhere in the order of 15 to 30 percent. However, part of these costs can be offset by reductions in transportation and boiler operation expenses.

Turning now to nitrogen oxide control, and later I'll mention combined sulfur oxide and nitrogen oxide control, we've estimated that about 20 million tons of nitrogen oxides were emitted from stationary and mobile sources in 1981. Coal-firing and utility boilers accounted for about 30 percent of these emissions. In addition to the relationship of NO_x to acid deposition, nitrogen oxides have been determined to be a criteria pollutant having adverse human health effects and a contributor to the formation of photochemical oxidants, another criteria pollutant.

For existing coal-fired boilers, one of the more effective NO_x reduction techniques is to reduce the level of excess air supplied to the burner. This relatively inexpensive technique, referred to as low excess air firing, can reduce NO_x emissions by as much as 50 percent and has the side benefit of, at least in some cases, increased boiler efficiency. However, all boilers do not have sufficient flexibility to achieve this level, and, in fact, this is a relatively limited technology.

A variation of this low excess air approach is to operate the lower levels of burners at the lowest excess air level possible and to provide additional air, sometimes referred to as over-fire air, in the upper regions of the boiler. EPA field tests on 22 pulverized coal-fired boilers utilizing these combustion modifications resulted in NO_x emission reductions ranging from 12 to 62 percent. Average for all these runs was about 37 percent. Capital costs averaged only about $2 a kilowatt, a relatively small expense.

The major thrust of EPA's low NO_x reduction research program is to develop the technology for new low NO_x burners which may either be retrofitted to existing boilers or incorporated into new designs. These burners reduce NO_x by delaying the rate of mixing be-

tween the fuel and the air, thereby limiting the availability of free oxygen in the initial burning process.

First generation technology has already been incorporated into the commercial application and was the basis for reducing the new source performance standard for NO_x in 1979 for coal-fired utility boilers. Capital costs are estimated to be about $5 a kilowatt.

While conventional combustion modifications have been used to achieve emissions below NSPS levels, we at EPA are exploring advanced combustion techniques to reduce NO_x even further. The most promising technique appears to be second generation, low nitrogen oxide burners.

Two such concepts developed under EPA sponsorship are the distributed mixing burning, for wall-fired boilers, and the rich fireball, for tangentially-fired boilers. Experimentally, these techniques have shown the potential to reduce NO_x emissions by more than 50 percent from current NSPS levels for pulverized coal units. The distributed mixing burner has been installed on an industrial boiler, about—it's a utility boiler, but it's industrial size, about 22 megawatts—and has achieved up to about 70 percent NO_x reductions under optimal conditions. The rich fireball, installed on a 400-megawatt, full-scale boiler, has achieved up to 50-percent reduction in NO_x emissions in long-term operation. We hope for new boilers that NO_x reductions greater than 50 percent from the current new source performance standards should be achievable.

I have separated out the category of combined sulfur oxide/nitrogen oxide control, because in addition to controls dealing separately with these pollutants, several important technologies simultaneously remove SO_x and NO_x In general, these combined technologies are developmental and not fully commercialized. Such technologies include limestone injection through multistaged burners—a retrofittable technology—and fluidized bed combustion, which we've heard quite a bit about so far, a complete coal combustion system generally for new applications.

Several advanced combined flue gas treatment technologies are being developed by DOE, and I think we'll hear more about them in the next presentation. These include an electron beam technique and an advanced copper oxide reactor, both of which simultaneously remove SO_x and NO_x from flue gases.

Of these combined technologies, LIMB may be most applicable to acid deposition control since it could be retrofitted at relatively low cost. This technology evolved from EPA sponsorship of low NO_x burners, which we've previously discussed. It was recognized that the conditions which resulted in reduced NO_x might also promote capture of sulfur compounds by adding sorbents, such as limestone.

Application of the technique to boilers could be a relatively simple and inexpensive way to reduce SO_2 emissions. Preliminary estimates indicate capital costs of less than 20 percent of those of scrubbers, although, of course, they don't achieve as much sulfur oxide removal. With increased interest in low cost control technology for acid rain precursors, in 1981, fairly recently, the EPA launched the LIMB R&D program for pulverized coal boilers. The primary objectives of the program are:

For retrofit, to remove 50 to 60 percent of both SO_x and NO_x from uncontrolled levels.

We also hope to realize cost reductions of between $50 to $150 per kilowatt less than alternative technologies such as flue gas desulfurization.

It should be noted that the research program to date has shown removal efficiencies ranging from as low as 20 percent to as high as 80 percent, depending upon factors such as coal type, alkali selected, and boiler conditions. We still have quite a bit of research to do before this would be considered commercial or even near commercial technology.

The recent EPA 1984 appropriations bill supplemented the LIMB development program with an additional $5 million for a total of about $9 million in extramural funding. The R&D program over the next 2 years will provide results that will allow, we hope, commercialization of the LIMB technology.

In conclusion, I would like to thank you for this opportunity to comment on several forms of existing and emerging technology which could be useful in acid deposition control. I believe technology is currently available for high levels of sulfur oxide control, albeit at high costs. The EPA research program is seeking to assist the development of additional acid deposition precursor controls for both sulfur oxides and nitrogen oxides, which will hopefully be effective, yet at substantially lower costs. However, the practicality of such technology has not been determined.

Thank you.

[The prepared statement of Dr. Frank Princiotta follows:]

STATEMENT OF

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BEFORE THE

JOINT HEARING OF THE SUBCOMMITTEE ON
ENERGY DEVELOPMENT AND APPLICATIONS
AND THE
SUBCOMMITTEE ON NATURAL RESOURCES,
AGRICULTURE RESEARCH AND ENVIRONMENT
OF THE
COMMITTEE ON SCIENCE AND TECHNOLOGY
U.S. HOUSE OF REPRESENTATIVES

SEPTEMBER 20, 1983

Good Morning, Mr. Chairman, and members of the Subcommittees. My name is Frank Princiotta. I am the Director of the Industrial Environmental Research Laboratory of the Environmental Protection Agency at Research Triangle Park. I appreciate the opportunity to meet with you and discuss several forms of existing and developing technology designed to control sulfur oxides and nitrogen oxides which, based on our current state of knowledge, appear to be the major pollutant precursors to acid deposition. DOE will focus upon technologies that they are developing.

Emissions from fossil fuel power plants have been the focus of attention for possible approaches to controlling the release of acid precursors, because nationally these sources produce a major share of the sulfur dioxide and a sizeable portion of nitrogen oxide emissions. In addition, other activities such as smelting, industrial combustion, transportation and space heating, when viewed together, can contribute significantly to

the atmospheric loading of sulfur and nitrogen acid precursors. My statement today will focus upon coal-fired utility boilers because our knowledge regarding those sources is more complete. I will also discuss those technologies capable of being retrofitted to existing boilers since control of such sources may be a major part of any future effort to control acid deposition precursors.

Table 1, attached, summarizes the major man-made acid deposition precursor pollutants, the major industrial sources, applicable controls and a brief explanation of their operation. Table 2, attached, focuses on coal-fired power plants, major emitters of both sulfur and nitrogen oxides. The table summarizes performance, commercial status, costs, and applicability for available and emerging controls. The following discussion will elaborate on the state-of-the-art and the Federal research program relating to the two major acid deposition precursor pollutants.

Sulfur Oxide Control

Currently, about 25 million tons of SO_2 are emitted annually into the atmosphere by various utility and industrial sources in the United States. In addition to being an acid deposition precursor pollutant, elevated sulfur oxides concentrations have also been associated with certain adverse health effects, visibility deterioration and material corrosion. These impacts have been considered in the setting of ambient air quality standards. The main approaches to reducing emissions of sulfur oxides from coal combustion other than switching to low sulfur fuels are removing sulfur from the coal, capturing the sulfur oxides during the combustion process, and removing or scrubbing the sulfur oxides from the combustion of flue gas. Of these methods, flue gas scrubbing (also called flue gas desulfurization---FGD) is the only technology that is used commerically on

a wide scale at electric utility plants. To date, more than 100 FGD installations scrub flue gas on about 44,000 megawatts of electrical generating capacity. If we add the planned FGD capacity, we expect over 200 utility FGD units to control over 100,000 megawatts of generating capacity in the United States by the turn of the century.

FGD processes can be classified as those that produce a waste product versus those that produce a saleable product such as sulfur or sulfuric acid. Almost all current and planned FGD installations are the waste producing type because these are generally the least expensive processes. The most common FGD processes that produce a waste product are those scrubbers employing lime or limestone as the alkali reagent. This reagent reacts with the acidic SO_2 yielding insoluble calcium salt waste. The processes that produce saleable products are more complicated and expensive. The cost of the current FGD technology is high in terms of total dollars and represents the most expensive air pollution control device for coal combustors. Capital costs for FGD retrofitting are typically $200 to $300 per installed kilowatt.

Since the early 1970's, EPA's research program has played a crucial role in upgrading the performance and reliability of the lime and limestone scrubbing technology. EPA has recently demonstrated the ability of organic acid additives to further improve performance and reliability over state-of-the-art limestone scrubbers. At EPA research efforts continue to seek lower cost FGD processes with particular emphasis on a spray dryer FGD system which offers the potential for significantly lower capital costs than corresponding wet systems.

Physical coal cleaning represents an alternative and/or supplemental sulfur oxide control approach. Coal is physically cleaned--a process sometimes called washing--by crushing and utilizing separation equipment to remove some of its mineral impurities, including inorganic sulfur. Conventional physical coal cleaning processes can reduce the potential sulfur emission from the combustion of coal typically by only 10 to 30 percent, depending on the specific coal.

Approximately 30 percent of all utility coals are currently cleaned. Curent practices reduce the potential annual SO_2 emission from coal production by 1.8 million tons. Conventional coal cleaning processes increase the cost by 15 to 30 percent. However, a part of these costs are offset by reductions in transportation and boiler operation costs.

Nitrogen Oxide Control

EPA recently estimated that about 20 million tons of nitrogen oxides were emitted from stationary and mobile sources in the United States in 1981. Coal-firing and utility boilers accounted for nearly 30 percent of these emissions. In addition to the relationship of NO_x to acid deposition, NO_x has been determined to be a criteria pollutant having adverse human health effects and a major contributor to the formation of photochemical oxidants, another criteria pollutant.

For existing coal-fired boilers, one of the more effective NO_x reduction techniques is to reduce the level of excess air supplied to the burner. This technique, referred to as low excess air firing, can reduce NO_x emissions by as much as 50 percent and has the side benefit of increased boiler efficiency. All boilers, however, do not have sufficient flexibility to achieve this level of emission reduction via low excess air.

A variation of the low excess air approach is to operate the lower levels of burners (utility boilers have multiple rows of burners located on the boiler's vertical walls) at the lowest excess air level possible and to provide additional air in the upper regions of the boiler. EPA field tests of 22 pulverized coal-fired boilers using these combustion modifications resulted in NO_x emission reductions ranging from 12 to 62 percent (the average reduction for all units was 37 percent). Capital costs average only about $2/kw.

The major thrust of EPA's NO_x reduction program is to develop the technology for new low NO_x burners which may either be retrofitted to existing boilers or incorporated into new designs. These burners reduce NO_x by delaying the rate of mixing between fuel and air, thereby limiting the availability of free oxygen in the initial burning process.

First generation burner technology has already been incorporated into commercial application and was the basis for reducing the New Source Performance Standard (NSPS) for NO_x in 1979 for coal-fired utility boilers. Capital costs are estimated at about $5/kw.

While conventional combustion modifications have been used to achieve emissions below NSPS levels, we are exploring advanced combustion techniques

to reduce NO_x even further. The most promising technique is second generation low NO_x burners.

Two such concepts developed under EPA sponsorship are the distributed mixing burner (for wall-fired boilers) and the rich fireball (for tangentially fired boilers). Experimentally these techniques have shown the potential to reduce NO_x emission by more than 50 percent from current NSPS levels for pulverized coal-fired units. The distributed mixing burner has been installed on an industrial-sized boiler (22MW) and has achieved up to 70 percent NO_x reductions under optimal conditions. The rich fireball, installed on a 400MW utility boiler, has achieved up to 50 percent reduction in NO_x emissions in long-term operation. For new boilers, NO_x reductions greater than 50 percent from current NSPS should be achievable.

Combined Sulfur Oxide (SO_x)/Nitrogen Oxide (NO_x) Control

In addition to separate controls for SO_x and NO_x, several control technologies simultaneously remove SO_x and NO_x. In general, these combined technologies are developmental and not fully commercialized. Such systems include limestone injection through multi-staged burners (LIMB)--a retrofittable technology, and fluidized bed combustion (FBC)--a complete coal combustion system for new applications.

Several advanced combined flue gas treatment technologies are being developed by the Department of Energy. These include an electron beam technology and an advanced copper oxide reactor, both of which simultaneously remove SO_x and NO_x from flue gases.

Of these combined control technologies, LIMB may be most applicable to acid deposition control since it could be retrofitted at relatively low cost. This technology evolved from the EPA sponsored low NO_x coal burner development work previously discussed. It was recognized that the condition which resulted in reduced NO_x might also promote capture of sulfur compounds by adding sorbents (e.g., limestone). Application of the technique to boilers could be a relatively simple and inexpensive way to reduce SO_2 emissions. Preliminary estimates indicate capital costs of less than 20 percent of the cost of scrubbers (flue gas desulfurization systems). With increased interest in low cost control technology for the major acid rain precursors (SO_x and NO_x), in 1981 the EPA launched the LIMB R&D Program for pulverized coal-fired boilers. The primary objectives of the program are: (1) for retrofit, 50-60 percent reduction of both SO_x and NO_x from uncontrolled levels; and (2) cost reductions of 50 to 150 dollars per kilowatt less than alternate technologies such as flue gas desulfurization. It should be noted that the research program to date has shown removal efficiencies ranging from 20 to 80 percent, depending upon factors such as coal type, alkali selected, and boiler conditions. The recent EPA 1984 Appropriations Bill supplemented the LIMB development program with an additional $5 million for a total of about $9 million in extramural funding. The R&D program over the next two years will provide results that will allow commercialization of the LIMB technology.

Conclusion

Thank you for this opportunity to comment on several forms of existing and developing technology which could be useful tools in acid deposition precursor control. In closing, I believe technology is currently available for high levels of sulfur oxide control albeit at high costs. The EPA research program is seeking to assist the development of additional acid deposition precursor controls for both SO_2 and NO_x which would be effective yet at substantially lower costs. However, the practicality of such technology has not yet been determined. If you have any questions I would be happy to answer them at this time.

TABLE 1 POTENTIAL CONTROL OPTIONS FOR ACID PRECIPITATION PRECURSORS

Major Acid Precipitation Pollutants Precursor Pollutants	Major Sources	Applicable Control Approaches	Principles of Operation
Sulfur Dioxide (SO_2)	- Electric Utilities; primarily high-S coal & oil power plants	- low sulfur fuel switching (commercial)	- Substitute naturally occurring low-S coal or oil.
		- physical coal cleaning (PCC) (commercial)	- Remove physically bound sulfur (pyrite) from coal via separation processes prior to combustion.
	- Industrial Fuel Combustion; primarily high-S coal & oil combustion	- flue gas desulfurization (FGD) (commercial)	- React acidic flue gas with alkaline liquids yielding either a throwaway or saleable sulfur compound product.
		- limestone injection multistage burners (LIMB) (developmental)	- Inject limestone in boiler through low-NO_x burners to react with sulfur gases to produce a throwaway product.
Nitrogen Oxides (NO_x)	- Electric Utilities; coal, oil, and gas fired	- low excess air (commercial)	- Minimize air used for combustion to reduce NO_x production from air N_2.
		- staged combustion (commercial)	- Add overfire air ports to allow fuel-rich early combustion conditions to reduce NO_x to elemental N_2 and O_2.
	- Industrial Fuel Combustion; coal, oil and gas boilers and process heaters	- low NO_x burners (1st gen. commercial; 2nd gen. burners in demo phase)	- Replace old burners with specially designed burners to maximize fuel-rich early flame conditions for NO_x reduction.
		- flue gas cleaning (FGC) (commercial in Japan)	- Treat flue gas with NH_3 in selective catalysts to reduce NO_x to N_2 and O_2.
	- Stationary Engines; large diesels, gas turbines, and IC engines	- catalytic reduction (demo phase)	- Utilize combustion gas catalytic systems to reduce NO_x to N_2 and O_2 either with NH_3 (fuel-lean engines) or w/o NH_3 (fuel-rich engines).
		- combustion modification (demo phase)	- Modify engine to encourage staged combustion.
	- Mobile Sources; autos and trucks	- exhaust gas recirculation (commercial since 1972)	- Recirculate exhaust gas back to cylinders to lower temp. and lower oxidation of air N_2 to NO_x.
		- three-way catalyst (commercial since 1982)	- Special tailpipe catalytic system to reduce NO_x to N_2 and O_2.

TABLE 2 RETROFIT CONTROL TECHNOLOGIES FOR COAL-FIRED UTILITY BOILER

Process	SO_2 and NO_x Removal Efficiencies	Status	Estimated* Capital Costs ($/kW)	Applicability	Problems
Flue Gas Desulfurization (lime or limestone scrubbing)	90% SO_2 Reduction	Commercial	200 to 300	Most large utility boilers	- High costs; especially for difficult retrofits.
Physical Coal Cleaning	20 to 30% SO_2 Reduction	Commercial	40 to 50	Centralized cleaning facilities can allow broad applicability	- Limited SO_2 removal; especially for difficult to clean coals. - Large quantities of pyrite solid residue for disposal.
Low-Sulfur Fuel Switching	60 to 80% SO_2 Reduction	Commercial	26 to 61	Most utility boilers	- Boiler derating sometimes required. - Potential displacement of locally mined coal.
Low-NO_x Burners (second generation)	60 to 70% Reduction	Early Commercialization	2 to 7	Most modern utility boilers	- NO_x removal only; no SO_2 removal.
Limestone Injection Multistage (Low NO_x) Burners (LIMB)	50 to 60% SO_2 Reduction Goals 50 to 60% NO_x Reduction Goals	Developmental	27 to 37	Research goal: Most utility boilers	- Questions must be resolved regarding performance and boiler reliability.

*Based on 300 MW, 3% sulfur coal, and Dec. 1982 dollars.

Mr. LEWIS [acting chairman]. Thank you, Dr. Princiotta.

Without objection, Dr. Princiotta, your testimony, and also yours, Mr. Mares, will be made part of the record.

Mr. Mares?

STATEMENT OF JAN W. MARES, ASSISTANT SECRETARY FOR FOSSIL ENERGY, AND ACTING DIRECTOR FOR POLICY, PLANNING AND ANALYSIS, U.S. DEPARTMENT OF ENERGY

Mr. MARES. Thank you, Mr. Chairman.

As the committee requested, I focused most of my statement on the technology of coal, specifically those emerging technical concepts that could play a role in reducing the release of suspected acid rain precursors in the future.

Before describing some of these technologies, however, I want to emphasize a couple of key points that are contained in the opening two pages of my statement.

First, there is little question that manmade contributions of sulfur dioxide and nitrogen oxides play a role in the impacts of acidic precipitation. The question is, how much of a role?

As I've pointed out on the bottom of the first page of my statement, the extent of damage done by acidic deposition and the rate at which it occurs are still uncertain.

These are key uncertainties for which we must have better answers before we can say with any degree of assurance what impact new—or, for that matter, existing—control technologies will have on the occurrence and impacts of acid rain.

The uncertainties still confronting the scientific community were underscored, I believe, Mr. Chairman, in the testimony of Dr. Christopher Bernabo, which this committee heard last week. Dr. Bernabo, as you know, is the executive director of the national acid precipitation assessment program. His testimony, as you recall, summarized the President's annual report—the program's annual report to the President and Congress which was issued in June. Let me depart for a minute from my formal statement to read a brief excerpt from his overview of that report. I quote:

> Acid deposition is probably the major contributor to acidification [in the Adirondack Lakes], but the rates of change and the factors controlling it are still poorly understood.
>
> At present, the extent of actual aquatic damages is not well established. The question of long-term trends in fresh water acidification also remains uncertain.
>
> Similarly, although long-term declines in high-elevation spruce are observed in New England, the relative importance of acid deposition versus other factors such as drought, disease, and other pollutants is not yet documented.
>
> The national program is speeding up investigations and analyses in order to determine the actual effects of acid deposition.

Now back to my statement: I might add that this task force will complete an assessment of current damages attributed to acid deposition by the end of fiscal year 1985. The 1985 date is based on realistic projections of how soon critical scientific information will be available.

In short, Mr. Chairman, scientists can quantify sources of the chemical compounds, like SO_x and NO_x, that contribute to acid rain, but today they are still unable to predict the environmental effects on any one region of the country that are due to sources from any other region.

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This was reaffirmed by the recently released National Academy of Sciences' report. I've included excerpts from the NAS report in my formal statement on page 2 because they reinforce the uncertainty that continues to exist within the scientific community regarding the impact of specific sources of acid rain precursors on specific receptor locations.

To put it simply, Mr. Chairman, we cannot say with any degree of certainty that reducing SO_x, NO_x, and other emissions from any one source, local or distant, will result in a specific reduction in acid rain, or its impacts, at any one receptor location.

On the other hand, we can say with some degree of certainty that trends in damage attributed to acid rain, if they exist, are measured in decades—I said measured in decades—not years. We are not on the edge of a precipice. We have the time to take rational, well-thought-out action, if called for, based on a solid base of scientific knowledge we expect to have available within the next few years.

A second point should also be stressed. The impression seems to exist in many quarters that the United States is doing little about the release of suspected acid rain precursors. Quite the contrary is true. As many members of this committee are aware, the United States has taken significant action, particularly in the last decade, to improve the quality of its air and water. The Clean Air Act is one of the most stringent environmental laws in existence anywhere in the world.

As Dr. Princiotta has just pointed out, some 44,000 megawatts of electrical generating capacity are now being scrubbed in the United States, with the total expected to rise to more than 100,000 megawatts by the turn of the century. I would add that these existing scrubbing units, along with other installed air pollution controls, represent more than $150 billion of investment on the part of American industry since the early 1970's.

As a result, emissions of sulfur dioxide have decreased—I repeat, have decreased—by 15 to 20 percent during the last decade. In all likelihood, beginning sometime in the 1990's, or perhaps around the year 2000, particularly in the northeast quadrant of the United States, these emissions will begin a second significant decline that will continue for the foreseeable future.

I would call your attention to figure 4 of the EPRI testimony this morning which shows that trend going downward.

The decline will take place as our pre-1971 utility power plants are retired and replaced or modernized using newer, cleaner technology, such as we've discussed here this morning. It will happen even if the Nation's GNP is high and current air quality standards are retained. If the standards are changed to reflect the emergence of these newer, more efficient control technologies, such as those that were described this morning or I'll describe that we're working on, the decline in sulfur emissions could be even more pronounced.

This is an important point, Mr. Chairman, so let me stress it again. Even under existing legislation, as older plants are replaced by newer, cleaner ones, emissions of sulfur dioxide will, in all likelihood, begin to decline significantly, starting perhaps before the

year 2000. This is especially significant given our knowledge that damage from acidic precipitation is at worst a function of decades.

Beginning on page 4 of my statement, I've outlined some of the promising technologies that could play a significant role in controlling the release of acid rain precursors from utility and industrial energy sources. While most of our research is focused on emissions of SO_2, NO_x, and particulates, the technologies being developed may also be capable of controlling some of the other chemical species thought to be important in acidic deposition. These include primary sulfates and volatile organic compounds.

As you can see, I've organized the description of these technologies by specific points where they would be implemented in coal's fuel chain, from the mine mouth to the plant stack. I would like to spend just a couple of minutes summarizing some of the new technologies at each of these points.

Precombustion coal cleaning may play an increasingly important role. As I've stated on page 5 of my testimony, the current new source performance standards allow credit to be given for any sulfur removed before combustion. However, today's coal cleaning, by itself, is not sufficiently effective to remove the 70 to 90 percent of the sulfur required to be removed in most coals.

Much of the limitation has to do with the nature of the sulfur itself. Coal contains essentially two types of sulfur, pyritic and organic.

Perhaps as much as 40 to 70 percent of the sulfur in some coals is pyritic sulfur; that is, it exists as physically distinct particles within the coal structure. All of the 1.8 million tons of potential sulfur emissions that Dr. Princiotta mentioned in his statement as being removed today by coal cleaning are of the pyritic form.

We believe it may be possible, through a combination of fine grinding and improved physical cleaning techniques, to increase the effectiveness of today's coal-cleaning technology.

On page 6 I've described some of the research goals for our coal preparation program. Current physical coal cleaning—coal washing, for example—removes between 33 and 50 percent of the pyritic sulfur in coal. We believe it may be possible to boost this effectiveness to 90 to 95 percent, equivalent to roughly 60 to 65 percent removal of the total sulfur in some very high-sulfur coals.

However, physical coal cleaning alone will not produce a compliance coal capable of meeting the NSPS standards, although it might enable some older plants to meet State implementation standards. To achieve the sulfur reduction levels currently mandated by the NSPS, organic sulfur—that's the sulfur chemically bound to the coal structure—must also be removed. This will require new, chemical coal-cleaning technologies, technologies that chemically break the bonds between the organic sulfur and the carbon atoms in the coal.

The chemical technologies described on page 6 of my statement, the TRW Gravimelt process and the GE Microwave approach, both appear capable of producing a low-sulfur, low-ash coal that can meet the current NSPS standards. Research is still needed, however, to bring costs to within a reasonable range and verify that the processes can be operated effectively in a continuous manner.

The primary goal of the Department's coal-cleaning program is to develop the technology base so that coal could be used as a substitute for oil and gas in existing applications. An important spinoff, however, could be new technology to clean high-sulfur coal for use in existing coal-fired powerplants. This would permit a plant to reduce SO_2 emissions without having to switch sources of coal.

Let me skip the combustion process, for the moment, and move to the end of the fuel chain; namely, the cleanup of the flue gas. Dr. Princiotta touched briefly on the existing and emerging technologies in this area. On pages 7 and 8 I have referenced some of the commercial alternatives to conventional or so-called wet scrubbing. The Government has played an important role in the early advances of concepts such as the spray dryer, and now the initiative has switched to industry. You will recall that was discussed in the previous panel, the spray dryer effort that we funded and that is being demonstrated right now at the Argonne Laboratory in Chicago using high-sulfur coal.

In turn, our R&D program has now been refocused on a more advanced scrubbing concept, one capable of removing from 90 to 95 percent of both SO_2 and NO_x emissions simultaneously. This technology could be commercially available within the late 1990's.

Other significant advances are being made in the combustor itself, particularly in the area of fluidized bed combustion which I've described on page 9.

Those of you who have heard me testify on this subject before will recognize a familiar statement: I believe that the fluidized bed coal combustor is one of the "success stories" of cooperative Government-industry R&D. Throughout most of the 1960's and 1970's, Government nurtured the idea of a combustion system that would burn a wide variety of coals and at the same time remove sulfur inside the boiler as well as minimize the formation of NO_x. When the technology appeared sufficiently promising, private industry took over.

Today more than 60 commercial atmospheric fluidized bed combustors are either operating, under construction, or on order. Every major boiler manufacturer either offers a fluid bed unit for sale or is developing a system for eventual marketing. And I might point out that Members of the Congress do not have to travel far to see an operating fluidized bed unit. One is about to enter its fourth year of operation at Georgetown University here in the District.

In the electric utility area, the private sector—led by EPRI and TVA—are moving forward to develop a large-scale fluidized bed demonstration unit that is expected to be the final development step before the technology becomes commercially available for powerplants in the 1990's. This initiative is described on page 10 of my formal statement and, as you know, there was a lot more discussion about that earlier—in the earlier panel.

In the Department's program we are concentrating on more innovative fluidized bed approaches that could improve the competitiveness of smaller industrial boilers. Oil and gas are still more economical in some of the smaller boiler applications, compared to fluidized bed, but if we can bring costs down by 25 to 30 percent—and that is our goal—fluidized bed systems would be able to compete favorably in the smaller boiler market.

We are also developing, in concert with both international and domestic research groups, the pressurized fluidized bed approach. This technique has the additional advantage of boosting powerplant efficiency through a combined cycle arrangement of both gas and steam turbines.

There are several other advanced coal technologies which have the potential of reducing the emissions of SO_x and NO_x in power-generating applications. I've listed a brief description of these concepts beginning on page 12. I won't use the committee's time to describe them here, although I or my staff could provide additional explanations or answer questions at the appropriate point or for the record.

The concluding pages of my statement, Mr. Chairman, deal with the budget totals for the activities I've described plus a discussion of the Department's involvement in the interagency acid rain research program. I might interject right now the chart you have over here, if it purports to be my program, it's false. I spend already on coal cleaning, NO_x and SO_x removal from flue gas, and coal preparation $18 million in 1983, and that shows less than that. So I don't know where you got the data, but it does not apply to my program.

I would point out that, just as the interagency effort involves a number of Federal agencies, each with their own expertise, several offices are involved within the Department of Energy, each bringing a diversity of knowledge, experience, and perspective.

In summary, let me stress that the Department places a high degree of emphasis on new technologies that can meet or exceed current NSPS standards in an economical manner.

Given the age of many existing powerplants, plus expectations for growth in energy demand, we project a significant opportunity becoming available around the year 2000 for the introduction of a variety of advanced technologies. This timing is consistent with the potential applicability of technologies expected to emerge from our R&D program.

We are actively involved in a research effort across the entire spectrum of Federal acid rain activities, from understanding better the scientific uncertainties to developing a new generation of technologies that could control the release of acid rain precursors more effectively. We are confident, Mr. Chairman, that our control technology program can be fashioned to be responsive to acid rain control needs.

That concludes my statement, Mr. Chairman. I'd be pleased to answer any questions that you may have.

[The prepared statement of Mr. Mares follows:]

Testimony

of

JAN W. MARES
Assistant Secretary for Fossil Energy
and
Acting Director for Policy, Planning & Analysis
U.S. DEPARTMENT OF ENERGY

Mr. Chairman and members of the Committee:

I am pleased to represent the Department of Energy at today's hearing and to join with the Committee in discussing the issue of acid rain and its implications for fossil energy research and development.

Acid precipitation has become an intense environmental issue with strong financial and social overtones. The implications of the current debate are neither confined to regional boundaries nor restricted by national borders.

The phenomenon of acid rain is not new; it has been active for hundreds of millions of years. Sulfur oxide compounds (SO_x), one of the so-called "precursors" of acid rain, have long been released to the atmosphere from natural sources. Oxides of nitrogen (NO_x), another suspected contributor to acid precipitation, are produced from such natural sources as lightning and organic decomposition. What is new, however, from a geological standpoint is the world's large-scale burning of fossil fuels -- in power plants, in homes and industries, and in our transportation sector. Today on a worldwide basis, for example, the level of sulfur released by human activity is about the same as is put there naturally. Industrialization and agricultural practices also account for a significant amount of the atmospheric nitrogen oxide content, although estimates of the relative contribution vary widely. Yet, because these human activities are concentrated in a relatively small area of the globe, the manmade addition of acid rain precursor pollutants to the atmosphere in some regions may exceed those of nature by 10 to 20 times. There is little question that these anthropogenic contributions play a role in the impacts of acid precipitation; the question remains "how much of a role?"

For example, the extent of damage done by acid deposition to aquatic and terrestrial ecosystems and the rate at which it occurs are still uncertain. Sources of the chemical compounds that contribute to acid precipitation can be pinpointed and quantified, but it is still impossible to predict the environmental effects on one region of the country due to sources from any other region.

As stated in the recently released study by the National Academy of Sciences' National Research Council Committee on Atmospheric Transport and Chemical Transformation in Acid Precipitation:

> "The relative importance of deposition at specific sites of long-range transport from distant sources as compared with more direct influences of local sources cannot be determined from currently available data or reliably estimated using currently available models."

Furthermore, the Committee concluded that:

> "It appears that the atmospheric processes in eastern North America lead to a thorough mixing of pollutants, making it difficult to distinguish between effects of distant and local sources."

In any discussion of acid rain, it is important to point out that the United States has not sat idly by during the last decade as the world's environmental consciousness has built. Since 1970, we have had in place one of the most stringent air quality laws in existence, the Clean Air Act and its subsequent amendments. The Environmental Protection Agency estimates that the U.S. has spent more than $150 billion during the last decade on air pollution controls. Nearly 100 flue gas desulfurization units, better known as "scrubbers", are now in place on about 44,000 megawatts of electrical generating capacity with more than 100 more either being installed or planned.

As a result, emissions of sulfur dioxide (SO_2) have decreased by 15-20 percent within the last decade. In all likelihood, these emissions may actually begin a second significant decline, particularly in the northeastern quadrant of the U.S., starting within the next 10 to 15 years, as older high-emitting sources are replaced with cleaner plants. This downward trend is projected to occur even assuming a high Gross National Product and increasing power demand.

In addition to the Federal and state regulatory requirements already in force, the federal government, working with industrial and academic researchers, has conducted since the 1960s and 70s an aggressive research and development program to improve the technologies associated with the combustion

of fossil fuels. This program has the potential for restricting the generation and emission of suspected acid rain precursors. As the Committee requested, I will concentrate my remarks today on this aspect of the acid rain issue, with a specific focus on technologies applied to the combustion of coal.

THE CURRENT REGULATORY FRAMEWORK

Control of SO_x/NO_x emissions from coal-fired boilers is mandated for new installations larger than 250 million Btu/hour thermal input by the New Source Performance Standards (NSPS). These standards were issued by the Environmental Protection Agency in 1971 and set an absolute ceiling on SO_x emissions at 1.2 lb/million Btu (of heat input) for all plants constructed or modified after August 1, 1971. In 1977, the Clean Air Act was modified and regulations were adopted to require coal-burning electric utilities built since Sept. 18, 1978, to remove from 70-90 percent of the original sulfur content in the "run-of-mine" coal, the exact value being a function of the type of coal utilized (for most eastern coals, removal requirements are closer to 90 percent than 70 percent). NO_x emissions are controlled to a ceiling of 0.5-0.6 lb/million Btu, again depending on coal type.

The release of sulfur and nitrogen-based compounds is generally expected to be reduced in areas where older power plants are replaced by newer units that must conform to these tightened NSPS emission requirements. However, the rate of replacement -- and consequently the attendant reduction in acid rain precursor emissions -- is a complex and uncertain issue. The decision to replace a plant is essentially an economic one; e.g., generally occurring when annual operating and maintenance expenses for the old plant outweigh the costs of a new plant. As a result of inflationary trends plus the costs of emission controls for new plants, there is a growing tendency to continue operating existing plants beyond their original 30-40 year design lifetimes.

Therefore, the research and development of new environmental control technologies must not only take into account concepts that limit the emissions

from new power generating sources but also potential retrofit technologies that could reduce the emissions from existing, older plants that may be pushed beyond their originally designed life expectancies.

CONTROL TECHNOLOGIES

Several control technologies are now available to reduce SO_2, NO_x, and particulate emissions. These technologies, as well as some of the newer concepts, may be capable in varying degrees of controlling some of the other chemical species thought to be important in acid deposition, such as primary sulfates ($SO_4^=$) and volatile organic compounds.

Reductions in SO_2 emissions can be achieved by lowering the sulfur content of the fuel through pre-combustion coal cleaning or by employing technologies that remove sulfur in the boiler or from the post-combustion flue gas.

NO_x emissions are a function of the combustion conditions as well as the nitrogen contained in the fuel. When air is heated to temperatures of more than 2,800 degrees F, the nitrogen and oxygen in it combine to form NO_x. Since nitrogen is also a significant component of coal, additional NO_x is formed when the so-called "fuel bound" nitrogen combines with oxygen during combustion. As a result, NO_x emissions from coal can be as much as twice the NO_x emissions of other fossil fuels.

The release of NO_x compounds from coal-fired utility boilers (which account for about one-third of all manmade NO_x emissions in the U.S.) can be controlled by modifying the combustion process or capturing the pollutant as it is released in the flue gas. Changing combustion conditions in existing facilities to reduce NO_x may be limited, however. Many utility boilers need high flame temperatures to operate efficiently both to achieve complete coal combustion and to facilitate removal of ash in a molten state. This contributes to NO_x formation; however, changing firing temperatures

would adversely affect boiler operations, and not all boilers have sufficient flexibility to achieve lower NO_x emissions by reducing combustion temperatures.

An important consideration in assessing some of the NO_x control technologies, such as low excess air and selective catalytic reduction, is the possibility that these approaches could actually increase volatile organic compound and sulfate emissions which might also be important factors in acid rain. Therefore, understanding the role that these species play in acid deposition becomes increasingly important in determining which control strategies are potentially most effective.

Pre-combustion cleaning: A minimum of 70 percent SO_2 capture is required for all coal-burning boilers that are subject to the current revised utility NSPS. Since the basis for determining SO_2 removal is the original Btu and total sulfur content of the coal as it comes from the mine, credit can be given for any sulfur removed before combustion. Today, about one-third of all coals intended for utility power plants are cleaned. Conventional coal cleaning alone, however, is not sufficient to remove the 70-90 percent of the sulfur content required of most coals.

Research sponsored by the Energy Department is attempting to develop a solid technological base for improving the capability to clean coal before it is burned. Not all coals will require the same amount of coal cleaning, but one target of this research program is to develop the capability to remove as much as 90 percent of the sulfur and up to 95 percent of the ash from some high-sulfur, high-ash coals. Results of several laboratory and small-scale tests now give us confidence that these levels can be attained, although costs must still be lowered to a reasonable range and the processes verified in systems designed for continuous operation.

To achieve this quality, coal may require grinding to sizes much finer than are common today, in some cases to particles roughly 1/1000th of an inch in diameter (finer than talcum powder). This releases greater amounts

of mineral matter making sulfur and ash removal easier and more effective. Grinding to fine sizes, however, is expensive because of the large amounts of energy required. A major focus of the Department's coal preparation program, therefore, is to develop various energy-efficient approaches to achieving the needed size reductions, including the use of high-frequency ultrasonic grinding devices, for example.

Following grinding, ash and sulfur can be removed in one of two ways -- physical or chemical coal cleaning -- or a combination of both. Conventional physical cleaning, which relies on properties such as density, surface characteristics, and magnetic susceptibilities, can theoretically reduce total sulfur and ash levels of some bituminous coals by an average of 10-40 percent. The sulfur removed is essentially "pyritic" sulfur ("pyritic" sulfur occurs as finely dispersed, physically distinct particles and makes up only a portion of the total sulfur in coal). The Department's research program is funding the development of improved physical techniques that could remove up to 90-95 percent of coal's pyritic sulfur (which would be equivalent to at least 65 percent removal of the total sulfur in very high-sulfur coals), compared to the 33-50 percent pyritic sulfur removal effectiveness of conventional physical coal cleaning.

For most U.S. coals, physical cleaning alone will be unable to produce a compliance coal capable of meeting the NSPS standards (although it might enable older plants in many locales to meet emission standards). To achieve the 90 percent total sulfur reduction level, organic sulfur (sulfur chemically bound to the coal) must also be removed. Chemical cleaning methods being developed with DOE funding either displace sulfur with basic alkali metals, such as sodium, calcium or potassium, or they oxidize the sulfur. Two alkali displacement concepts being pursued by the Department are the TRW Gravimelt process and the GE Microwave process. Both appear capable of producing a low sulfur, low ash, high-Btu coal product that can meet the current NSPS for SO_2 emissions.

While the primary goal of the Department's program is to produce a coal sufficiently clean to be used as a substitute fuel in applications for which oil or gas are presently used, a potential spinoff of this program could be

new technology to clean high-sulfur coal for use in existing coal-fired power plants. This would permit a power plant to reduce SO_2 emissions while retaining the same source of coal feedstock.

Post-combustion cleaning: During the past decade, a significant evolution in scrubbers has taken place within the power industry. In the mid to late 1960s, with initial promulgation of local sulfur dioxide standards, electric utilities use flue gas scrubbing principally as a last resort. During these early years, other less costly options for mitigating the effects of sulfur emissions existed, such as the use of low sulfur coal, dispersal by tall stacks, intermittent control by load or fuel switching, or coal cleaning. These alternatives, for the most part, were operations which were well understood by utilities, and equally important, did not require large nonproductive capital outlays. But with the promulgation of more stringent national sulfur dioxide control regulations, continuous SO_2 removal became a requirement rather than an option.

Flue gas desulfurization technology is capable of being retrofitted on many existing, stationary combustion sources, although it has been used mostly on new plants. The systems can achieve high SO_2 removal efficiency (70 to 90 percent); however, capital costs are high. A recent survey of major eastern electric utilities showed that the costs of retrofitting a scrubber to an existing power plant would range from $169 to $429 (1982 dollars) per kilowatt. (The wide range is caused by a variety of site specific factors, including the degree of modification required at the power plant, the costs of installing sludge disposal facilities, and how much redundancy the utility desires.) This is compared to a cost of approximately $1000 per kilowatt to build a new, pulverized coal-fired plant with a scrubber. In addition, the utility could have to divert up to approximately six percent of the plant's energy output to run the scrubbing process.

Of the variety of flue gas clean-up systems available today, more than 85 percent are the "wet" lime/limestone systems. The alkaline lime/limestone reagent reacts with the SO_2-laden flue gas to produce a residue of insoluable calcium salts. Disposing of this residue is a principal drawback of the

wet scrubber. When it leaves the power plant, the residue is a wet sludge that remains continually moist. The sludge is difficult to concentrate and must be stored in large settling ponds lined by clay or plastic to prevent seepage. During its lifetime, a typical 500-megawatt power plant with a wet scrubber is likely to require a sludge disposal pond of more than 400 acres and perhaps up to 40 feet deep. As much as 20 percent of the capital and 5-10 percent of the operating costs of a scrubber system typically are for sludge disposal.

Recently, some utilities have begun installing a refinement of the wet scrubber based on "forced oxidation" technology which converts the sludge at the power plant into a dry product that may have commercial value.

In addition, industry has taken the lead in developing a fast-growing alternative to wet scrubbing -- the spray-dryer system. A fine wet mist of dissolved or partly dissolved alkaline chemicals is still used to capture sulfur dioxide inside the system; however, the amount of moisture is controlled so that the hot flue gas is only partially saturated and the reaction products remain relatively dry. The solid waste can be caught and disposed of or maintained more easily than the wet sludge of more conventional scrubbers.

Spray drying has gone from pilot to commercial application in the last six years. At least 24 spray drying units have now been sold to industries and utilities in sizes of up to 860 megawatts. Most have been for facilities using western low-sulfur coal, although the concept has been shown to be economically applicable to higher-sulfur eastern coals. In 1981-82, pilot scale spray dryer tests at the Department's Pittsburgh Energy Technology Center demonstrated that better than 90 percent sulfur capture could be achieved from the flue gas of high sulfur coals. Since 1982, one of the first dry scrubbers in the world designed for high-sulfur coal combustion has operated successfully at the main steam boiler plant at the Department of Energy's Argonne National Laboratory outside of Chicago.

With these technical advances now taking place at commercial installations, the Department's flue gas cleanup program has refocused its research efforts on a new, more advanced generation of scrubbers capable of simultaneously removing SO_2, NO_x, and in some cases, particulates from the flue gas of coal combustion.

One approach would beam electrons through combustion gases that have been treated with either ammonia or an aqueous lime slurry. The electrons cause a complex series of chemical reactions to take place in which the sulfur and nitrogen oxides are converted to dry materials that can be removed by conventional techniques. Another concept would use copper oxide to remove SO_2 at temperatures also suitable for simultaneous nitrogen oxide removal with ammonia.

These advanced scubbing concepts are expected to be capable of reducing sulfur dioxide emissions by 90 to 95 percent, comparable to conventional lime/limestone scrubbers but with lower capital and operating costs and reduced energy penalities. In addition, they are being designed to reduce NO_x by a similar 90 to 95 percent, considerably in excess of conventional scrubbers which typically capture only 10 to 20 percent. Like the spray dryer, the simultaneous SO_x/NO_x removal systems generally produce dry products, some of which may have commercial value.

Clean-up during combustion: Coal-fired combustors have relied on essentially the same principles since the 1920s when pulverized coal was first used to generate steam. Industry has improved this technology so that today's boilers are larger, produce steam at much higher temperatures and pressures than half a century ago, and are more efficient in converting the energy in coal into steam. But the concept of grinding coal to very small sizes and blowing it with air into large furnace cavities, where the cloud of coal dust burns much like a fuel gas, is still the way most coal is burned today.

Within the past decade, however, a new concept in coal combustion has moved off the drawing boards and into the industrial market. Developed through a joint government/industry program, "fluidized bed combustion" today has become the first major advancement in modern-day coal combustion.

The major advantage of fluidized bed combustion is that 90 percent or more of the sulfur in coal can be removed inside the combustor, eliminating

the need for a scrubber. A suspended bed of limestone (or dolomite) inside the combustor is chemically changed into lime and absorbs the SO_2 before it escapes the boiler. The product is a dry, inert substance that is discharged with the ash. Also, the temperature required for the best sulfur capture is sufficiently low so that NO_x emissions are about half the New Source Performance Standards.

Capital costs of fluidized bed boilers are significantly higher than conventional oil- or gas-fired boilers (5-8 times); however, the lower fuel costs of coal and the elimination of post-combustion sulfur removal equipment significantly improve the economics of fluidized bed systems. Particularly at larger scales, the "lifecycle" costs of a fluidized bed combustion unit compare favorably to oil and gas firing.

Today, the atmospheric pressure version of the fluidized bed concept is commercially available for large industrial applications. Nearly every major boiler manufacturer either offers a fluidized bed combustor for sale or is developing a commercial system for eventual marketing. Approximately 60 atmospheric fluidized bed systems are either operating, under construction or on order. One example is the unit which now supplies steam for the campus of Georgetown University in Washington, D.C. The system, built as part of a joint government-industry-university development project, will soon be entering its fourth winter of operation.

The electric power industry has assumed the initiative in developing a utility atmospheric fluidized bed (AFB) power plant, building on the technology base developed by DOE and others. A prototype utility-scale system is being tested at the Tennessee Valley Authority's (TVA) $68 million, 20-megawatt (200,000 pounds of steam) pilot plant near Paducah, KY, and the Electric Power Research Institute has decided to proceed in partnership with TVA to design and construct a 160-megawatt demonstration power plant. If this demonstration unit proves successful, the nation's electric utilities would have the option of building new AFB power plants in the 1990s that could burn coal more efficiently while producing fewer SO_x and NO_x emissions than existing coal-fired plants.

The Energy Department is currently concentrating on more innovative fluidized bed concepts having the potential for making coal more competitive in smaller industrial boilers where oil or gas is now more economical. This effort is designed to reduce capital and operating costs by 25 to 30 percent over conventional AFB costs. In April 1982, the Department selected seven advanced fluidized bed concepts for initial evaluation. Bench scale engineering studies of the most promising of these concepts will be conducted in FY 1984.

In addition, the Department is continuing to sponsor the development of the pressurized fluidized bed concept which could make "combined cycle" operation -- a more efficient electrical generating approach employing both gas and steam turbines -- a viable option for future coal-fired power plants. When developed, the pressurized fluidized bed boiler could also become a candidate to replace existing boilers in a power plant to increase the plant's electrical output while decreasing its environmental intrusion.

Information is being collected from the 80-megawatt (thermal) pressurized fluidized bed (PFB) boiler in Grimethorpe, England, operated with funding from the U.S., the United Kingdom, and the Federal Republic of Germany. Federal funds are being used to complete the construction of the 13-megawatt (electric) fully integrated PFB pilot plant at Wood-Ridge, NJ., with the Curtiss-Wright Corporation having agreed to secure private funding for the plant's operation.

SO_2 emissions from a pressurized fluidized bed boiler are expected to be reduced from the level emitted from an atmospheric fluidized bed system, while NO_x emissions are projected to be about the same. The improved thermal efficiency of the pressurized fluidized bed unit (projected to be about 40 percent compared to the 33-35 percent efficiencies of conventional coal-fired plants with scrubbers) would further reduce the overall environmental burden because more energy would be generated per unit of fuel consumed.

To meet NO_x emission levels, many commercial facilities are already applying combustion modifications such as staged combustion (although no SO_2 reduction is achieved). Such modifications are relatively inexpensive compared to the capital costs of the boiler; however, the NO_x reduction potential is limited (10 to 30 percent below uncontrolled levels). Among the emerging control technologies, low NO_x burners are particularly effective and are relatively inexpensive in terms of capital and operating costs.

The Environmental Protection Agency (EPA) is developing several advanced generations of combustion process modifications, including a retrofittable burner configuration termed the Limestone Injection Multi-Stage Burner (LIMB) which has the potential for moderate SO_2 removal as well as NO_x removal from some boilers. Like many of the advanced combustion technologies, however, LIMB is still in a prototype developmental stage and may not be commercially available until the 1990s. DOE is maintaining close coordination with EPA's research efforts in combustion modification as well as in pre- and post-combustion cleanup R&D.

OTHER ADVANCED COAL TECHNOLOGIES

Several other emerging coal technologies have the potential of impacting the emissions of SO_x and NO_x in power generation applications:

Integrated Gasifier/Gas Turbine Combined Cycle: This technology is typified by the 100-megawatt Cool Water power plant now under construction at Daggett, California. When completed in 1984, the commercial-scale demonstration project will integrate the gasification of coal with the generation of electricity by a combination of gas and steam turbines.

The California site picked for the Cool Water project illustrates an important feature of this new power generating concept. Exhaust emissions of particulate matter, SO_x, and NO_x are expected to be well below Federal NSPS requirements for new power plants, as well as within the still tighter limits required by California law.

In the integrated gasifier/combined cycle approach, sulfur is effectively removed in the gas cleaning process to meet NSPS as well as to control turbine blade corrosion. Fuel bound nitrogen comes off in the form of ammonia which is washed out in the quench process or which can be catalytically decomposed in a hot gas cleanup process. Combustion modification can be used to control the formation of NO_x from the air. While this technology has the potential for emissions control well below NSPS, in actual practice it is anticipated that SO_2 emissions in the gases released from the sulfuric acid or sulfur recovery process will be at or just below the NSPS.

Within the Department, activities are underway to improve the gasification/combined cycle concept by developing more effective means to clean impurities from the hot gases exiting the coal gasifier.

Integrated Gasifier/Fuel Cell Combined Cycle: Unlike other coal systems, fuel cells do not rely on combustion and consequently do not produce the SO_x and NO_x emissions associated with burning fossil fuels; instead an electrochemical reaction generates electricity. Working with the gas and electric utility industry, the Department is developing fuel cell systems capable of operating on natural gas and coal-derived fuels with long enough lifetimes and sufficiently low costs to be economically practical and reliable.

While any fuel cell can be linked to a hydrogen-rich fuel source, the fuel cells capable of operating at higher temperatures -- such as the molten carbonate (1200 degrees F) and solid oxide (1800 degrees F) systems -- are expected to be more suited to integrated operation with a coal gasifier.

To prevent electrode poisoning, sulfur in the fuel source must be removed down to the parts per million range in a gas cleaning process. Emissions from the sulfur removal system should be analagous to the integrated gasifier/gas turbine case. Fuel bound nitrogen can be removed in similar fashion as in the gasifier/gas turbine system, and NO_x from the air should not be a problem given the relatively low reaction temperatures occuring in the fuel cell.

Magnetohydrodynamics (MHD): This concept, while still confronted by several development stages, offers the potential for reducing SO_x and NO_x emissions without the use of post-combustion cleanup technologies. SO_x is captured by the potassium seed required to render the hot combustion gases electrically conductive. Sulfur would be recovered in the form of sulfuric acid or elemental sulfur with SO_2 emissions in the resultant off gas controlled to NSPS levels. NO_x would be controlled through the use of combustion modification.

Like other advanced coal-fired options, MHD also offers improved thermal efficiencies, perhaps as high as 50 percent. Since a greater yield of useful energy is obtained from each unit of fossil fuel consumed, MHD could help achieve a reduction in environmental burden.

MHD, however, has several technical uncertainties well beyond the current state-of-the-art that must be addressed before utilities have sufficient confidence to move the technology into commercial development. In the environmental area, potentially difficult issues have not been addressed. For example, little is known about water pollution from the leaching of potassium salts from coal ash and refuse from the potassium seed regeneration wastes, nor is there much information on possible air pollution caused by potassium salt release from particulate control equipment.

Therefore, it may be more appropriate to focus research on answering these more fundamental scientific and environmental uncertainties associated with MHD rather than pursuing rapid large-scale engineering demonstration with its attendant high costs and risks.

BUDGET PROFILE

The budget levels for the relevant DOE/Fossil Energy programs described in this testimony are as follows:

(in millions of dollars)

Activity	FY1982 BA	FY1983 BA	FY 1984 Admin. Request	FY 1984 House Auth/Approp		FY 1984 Senate Approp[1/]
Coal Prep.....	$ 2.88	$ 8.00	5.00	8.00	8.00	8.00
Control Tech.						
Flue Gas.....	6.24	7.00	7.60	9.60	8.35	8.10
Gas Stream....	9.98	10.00	4.45	7.00	7.00	7.00
Cleanup Base..	5.28	2.90	0	3.20	3.20	2.00
Fluidized Bed Combustion ...	35.76[2/]	17.43	4.40	7.40	9.00	7.40
Fuel Cells	34.46	29.94	9.50	26.50	46.20	39.30
MHD.....	21.84	29.00	0	30.00	30.00	30.00

1/ Recommendation of the Senate Committee on Appropriations
2/ Offset by $10.5 million applied from previous fiscal years.

ACID RAIN RESEARCH PROGRAM — DOE INVOLVEMENT

In addition to sponsoring the development of new, more environmentally acceptable fossil energy technologies, the Department also participates in the Interagency Task Force on Acid Precipitation. Created by the Acid Precipitation Act of 1980, P.L. 96-294 (a component of the Energy Security Act of 1980), the Task Force includes 20 members, representing 12 Federal agencies, as well as four national laboratories and four Presidential appointees.

The DOE Office of Energy Research represents the Department on the Task Force, while DOE's Office of Fossil Energy heads a specific effort to investigate the role of manmade emissions believed to be important in acid rain formation (one of 10 "task groups" that make up the Interagency Task Force).

Overall, the Department plans to commit nearly $3.25 million to the nearly $27.5 million of research to be conducted by the task force in FY 1984. The DOE funding, which is provided by three offices including Energy Research, Fossil Energy, and Environmental Protection, Safety, and Emergency Preparedness, is directed toward the following task group areas:

Manmade Sources	$260,000	Aquatic Effects	$ 550,000
Atmospheric Processes ..	610,000	Terrestrial Effects ..	50,000
Deposition Monitoring ..	602,000	Assessments & Policy Analysis	1,175,000

In addition, the Department is involved in the Control Technology Task Group which is tracking all relevant control hardware development efforts and providing the appropriate agencies with guidance related to the National Acid Precipitation Assessment Program, and the International Task Group which is facilitating international cooperative research.

As part of its effort to provide an accurate and complete inventory of man's potential impact on the occurrence of acid rain, the Man-Made Sources Task Group completed in fiscal 1982 a preliminary study that suggested that both nearby petroleum combustion and local and distant coal combustion could significantly affect local acid deposition. The group is continuing to develop the models necessary to predict the cost and emission changes from utilities and industries based on various possible control strategies. By the end of FY 1984, the group is scheduled to have transferred to government computer systems a fully documented utility simulation model that will include data at the State level. Also in FY 1984, an industrial combustion emissions model will be completed, and work will continue to apply computer analysis methods to emissions from other, non-combustion industrial processes.

DOE is also heavily involved in the Assessments and Policy Analysis Task Group whose job is to integrate the results of all the research areas in a series of assessments that evaluate acid rain science. The group's initial major product is to be delivered in FY 1985 and additional assessments are to be released through 1989. In addition to contributing more than $1 million to assessment activities, DOE has committed personnel to perform assessment functions as part of an integrated EPA-DOE-national laboratories staff activity.

SUMMARY

Emissions from fossil fuel power plants have been the focus of attention for possibly controlling the release of acid precursors because nationally these sources produce a major share of sulfur dioxide and a sizeable portion of the nitrogen oxides. Other activities, however, such as industrial combustion and processes, transportation, space heating, and smelting, can all

contribute significantly to the atmospheric loading of sulfur and nitrogen acid precursors. All may be important (locally and/or regionally) to the formation of acid deposition, but their specific roles are still poorly understood.

Within the Department's Fossil Energy R&D program, a high degree of emphasis is placed on those combustion and advanced power cycle technologies that offer the potential for meeting or exceeding current NSPS standards in an economical manner. DOE energy projections indicate a significant opportunity around the year 2000 for the introduction of a variety of advanced technologies. This timing is consistent with the potential applicability of long-term technological advances expected to emerge from the DOE fossil energy R&D program.

Improved coal preparation technology, e.g., chemical coal cleaning, offers the potential for considerable reduction in SO_x emissions from existing plants. Research and development of advanced flue gas control technologies could lead to the economical and effective removal of SO_x, NO_x and respirable particulate matter for conventional coal combustion plants in a more efficient and cost-effective manner than is possible today. Several of these advanced flue gas technologies offer the prospect for economically retrofitting existing coal-fired plants. New combustion and power generation technologies, ranging from fluidized bed combustion to fuel cells, may offer options for new power plants to produce energy more cleanly from coal in the future.

Since the Department of Energy participates in the entire spectrum of the federal acid rain research and assessment effort, our control technology R&D programs can be fashioned to be responsive to acid rain control needs.

Thank you for the opportunity to present an overview of these activities.

Mr. LEWIS. Thank you, Mr. Mares.

Dr. Princiotta, what potential problems are foreseen in the development process for the lime-injection multiple burner?

Dr. PRINCIOTTA. Well, the real challenge—I say the real challenges in a successful demonstration of LIMB will include achieving our goals of 50 to 60 percent SO_2 removal efficiency at reasonable levels of limestone addition. We're reasonably optimistic this will be the case, but one does not want to put in more limestone than a certain critical level because then slagging, plugging, and other boiler-related problems could result.

Also, of course, the economics tends to look less favorable as you have to add more alkali for a given SO_2 removal requirement.

Mr. LEWIS. How about for industrial boilers? Is LIMB a potential application for that?

Dr. PRINCIOTTA. Yes; we—I should point out there are two classes of boilers that are important. One is the wall-fired class of boilers and the other are corner-fired or tangential boilers. Our work is much more advanced in the area of wall-fired boilers, but for both utility and industrial sources corner-fired or tangential boilers are important as well.

The answer, the fast answer is, yes, LIMB should ultimately be applicable to the larger class of industrial boilers. Especially we feel particularly optimistic about its applicability to wall-fired boilers. More work needs to be done for the corner-fired boilers.

Mr. LEWIS. When do you feel that LIMB would be ready for commercial use?

Dr. PRINCIOTTA. Well, assuming we do get a go-ahead on a demonstration program, the earliest such a program could probably be initiated would be about 1987. Assuming a year of successful operation, we're talking essentially about late this decade technology might be available for a larger-scale application. Perhaps by 1990 we feel comfortable in applying this to large numbers of boilers.

Mr. LEWIS. To what extent are you coordinating your research efforts of LIMB with Germany and other nations?

Dr. PRINCIOTTA. We have a very active program. In fact, we—I have personally worked with the UBA, the German EPA, in a cooperative program on their LIMB demonstration activity. As we heard earlier, they will next spring test a LIMB concept on a full-scale boiler on burning 1 percent black coal, and we are going to have our own contractor, our own observers, and we'll actually be taking data supplemental to the Germans. So we have a very closely coordinated program with the Germans, a very close relationship.

Mr. LEWIS. I see.

On page 6 of your testimony, you note that flue gas desulfurization processes that produce salable products are more complicated and expensive than those that are not. How much more expensive are these processes and how much more additional cost can be recovered—how much of the additional cost can be recovered by the sale of the product?

Dr. PRINCIOTTA. Probably the two most important regenerable processes that are commercial are the magnesium oxide process and the Wumerlaur process, and normally our cost studies suggest capital costs, depending upon the particular application, of some-

where between 20 and perhaps 40 percent higher than, let's say, a limestone scrubbing system. Typically, operating costs are also higher. Normally you need more electricity, more steam, and other ingredients to make this particular type of scrubber work. Some of that is offset, of course, by the recovered product. And, of course, depending upon the value of the recovered product, be it sulfur or sulfuric acid or some other product, you do get some benefit, but normally the value of the recovered product is a relatively small fraction of the total annual revenue requirements associated with that control device. It would take a huge increase in the value of sulfur products for this situation to turn around.

So, in summary, even assuming sale of the byproduct at the current market price, normally and generally the regenerable products are not competitive with the limestone throwaway systems. There are exceptions to that, though, in that if a utility has a particularly difficult task in disposing of the waste products and would have to incur a high cost in doing that disposal operation, it may be less expensive to go with the regenerable process.

Mr. LEWIS. Dr. Princiotta, the National Academy of Sciences, in its most recent report on acid deposition, concluded that there was no evidence for a strong nonlinearity in the relationships between long-term average emissions and deposition. A number of reputable scientists from several Department of Energy national laboratories disagreed both with the methods used to reach this conclusion and with the conclusion itself. Do you agree or disagree with NAS?

Dr. PRINCIOTTA. I am sorry, I am an engineer pretty much familiar with the control technology business, but I would be speculating in an area outside my expertise to comment on that particular question.

Mr. LEWIS. OK. Thank you.

Would you like to comment on that question, Mr. Mares?

Mr. MARES. Well, I am an engineer, but I will comment.

I have--my understanding is that four different national laboratories asked at least four, and in some cases six or seven, scientists at their area, at their laboratories that are doing scientific work in the area that is covered by the NAS. That means you've got a total of somewhere between 16 and 24 scientists. Each laboratory did an analysis that was submitted to your committee. Each laboratory challenged some aspects of that analysis.

My understanding is that in the paper that Dr. Calvert presented to your committee he, in his appendix, tried to evaluate that. He only referred to three of the somewhere between—or four, three of the 16 to 20-odd scientists who would evaluate it.

My sense is that the national labs have got some pretty good points that need to be thought about. The net effect is there is a lot of uncertainty. I think even the NAS agrees that there's a lot of uncertainty and that they were pushing somewhat the limits. I think that even was mentioned in your own—in the testimony before you last week.

So, yes, sir, I generally think that there is some real question about that conclusion.

Mr. LEWIS. Mr. Mares, in contrast to acidic or acidifying substances, what sources exist for neutralizing substances, including

ammonia, soil-related or cement dust, and alkaline particles from combustion?

Mr. MARES. Well, at the soil level there are the whole bunch of alkaline materials that are in the soil that are there for—that do neutralize acidic components. In the combustion side we are doing research, as mentioned by all of us here today, about adding limestone at various stages of the process in order to do that neutralization. The answer is there are both natural forms as well as the possibility and probability of adding alkaline agents to serve neutralization purposes.

Mr. LEWIS. The Acid Rain Peer Review Panel established by the Office of Science and Technology Policy recommended that additional steps be now taken to reduce emissions of sulfur compounds. Do you agree with this recommendation?

Mr. MARES. Well, the administration has not taken a position to date on that. I would like to read a pertinent part of that press release which they gave, which seems to be omitted. That is that they said, "We recommend that additional steps should be taken now which would result in meaningful reductions in the emissions of sulfur compounds in the atmosphere"—and this is the part that gets forgotten—"beginning with those steps which are the most cost-effective in reducing total deposition."

And they indicate specific things, and these were in your—in the hearing last week. They say, "Purely as an example, it may be useful to consider having fuel of different sulfur content during different seasons."

As other examples, intensifying coal washing.

I think that—the administration has not taken a position, but I think as people read that report they should make a full report and not take a sentence out of context, part of a sentence out of context.

Mr. LEWIS. I see.

Mr. MARES. I might make one other technical comment. I think that at least within our organization there's a very—there is a disagreement regarding one of the assertions about trends within the last 10 to 20 years. We think it's assertion 5 in that report. We think that it is more accurate—a more accurate statement would be over the last 10 to 30 years. We don't think the data substantiates the statement that's been presented there, and I would go back to my earlier comment that we think that the trends over this period are very uncertain and that if there is a rate of change it is very slow, perhaps spanning a period of decades.

Mr. LEWIS. Thank you.

Mr. Nelson from Florida?

Mr. NELSON. Thank you, Mr. Chairman.

On page 2 of your testimony, Mr. Mares, you state that emissions of sulfur dioxide have decreased by 15 to 20 percent within the last decade, and you state that within the next 10 to 15 years these emissions will begin another significant decline, especially as the older plants are replaced.

Tell me, legislatively we're looking at the Clean Air Act right now. If you take the present law under the Clean Air Act, will it accomplish those same results in sulfur dioxide reductions as compared with the proposed acid rain legislation?

Mr. MARES. Over time the current statute will.

Mr. NELSON. So over the next decade——

Mr. MARES. I think that's pretty well demonstrated in figure 4 of EPRI's testimony.

Mr. NELSON. All right. In addition to those reduced emissions of sulfur dioxides—and, by the way, let's go back to that figure 4. I haven't seen that. Would you describe that for me?

Mr. MARES. Well, rather than my trying to orally describe the chart, maybe one of the staff——

Mr. NELSON. All right. Let's go on, then.

Mr. MARES. Basically what it shows is that the trend of emissions from utilities of SO_x will be decreasing over time and that at least in whatever was the proposal—if I read the caption correct, it's retrofit control for acidic deposition reduced by 8 million tons between 1985 and 1990—that it reduces from the trend during a period of from 1985 to the year 2010. So that you're talking about, if I interpret the chart correctly, and I've seen one similar to this, that that particular proposal accelerates for a relatively short period of time in the context of our information about trends of a process of SO_2 emission reductions which is going to be taking place anyway.

Mr. NELSON. The reductions under the acid rain proposal reduce dramatically in the early years but they would cross—they would equalize out somewhere around 2010, 2015?

Mr. MARES. That's what that particular graph shows, yes, sir.

Mr. NELSON. Do you agree with that graph, figure 4?

Mr. MARES. Well, that's what the graph shows, yes, sir.

Mr. NELSON. All right. That was with regard to testimony by Kurt Yeager; is that correct?

Mr. MARES. Yes, sir.

Mr. NELSON. All right. Do you agree with that graph, figure 4? You said, "That's what the graph shows." Do you personally agree with the conclusions of that figure 4?

Mr. MARES. Yes, sir; that's what I testified, that over time the emissions under the existing statute are going to be reduced, and that particular proposal would accelerate a process which legally is already underway.

Mr. NELSON. All right. In addition to the reduced emissions of sulfur dioxides, are there additional benefits by coal cleaning?

Mr. MARES. Yes, sir. You eliminate the ash, which makes the combustion easier or more efficient. You've got a bit more flexibility there. Depending upon where the coal cleaning takes place, it may or may not reduce cost of transportation. The answer is yes, there are benefits above and beyond reduction of sulfur as a potential emitter from enhancing our extent and ability to wash coal or clean sulfur and ash from coal before it's combusted.

Mr. NELSON. Is there a connection between acid deposition in the northeastern United States and the high incidence of oil that is burned in that region?

Mr. MARES. I think there are a significant number of scientists that have—would say the answer to that is yes, that when you look at the meteorology and where plants exist that emit sulfur, sulfur that is deposited in the Northeast can come from a whole host of places. Nobody can assert with any degree of certainty exactly where it comes from.

Mr. NELSON. Might you put a percentage on the emissions of nitrogen oxides and sulfuric acid formed in catalytic converters?

Mr. MARES. Well, NO_x is certainly formed in the combustion of oil, gasoline, as well as in the burning of coal, and NO_x is a contributor to acidic deposition. NO_x comes from mobile sources as well as stationary sources. The answer is yes, it comes from a whole host of places.

Mr. NELSON. The question was, do you have an idea of how large a proportion is as a result of the mobile sources?

Mr. MARES. I'd rather answer that for the record, rather than guess at it, sir.

Dr. PRINCIOTTA. Perhaps I can answer that one. I think at this stage of the game approximately 40 to 45 percent of nitrogen oxide emissions are from the mobile source category, including cars and trucks and other vehicles.

Mr. NELSON. All right. Mr. Chairman, if I may, just a couple more?

You noted that DOE's Office of Fossil Energy heads up the interagency task force on acid precipitations, manmade emissions task group.

Mr. MARES. Right.

Mr. NELSON. What are the main research goals of that group?

Mr. MARES. That group is developing estimates of past, present, and future emissions of pollutants from manmade sources and then devising quantitative methods to estimate the effects and costs of possible Government regulation of those emissions. It is a research into data gathering, analysis, and projection.

Mr. NELSON. All right. With that data gathering and analysis, then, what is the long-term research objective?

Mr. MARES. Well, the long-term research objective of the programs that I've described try to identify ideas, develop them to the stage where they can be technically shown to work, that would remove sulfur and ash from coal before it's burned, so that it would be potentially either—could be used in existing powerplants or in place of oil in existing oil-fired facilities, to remove SO_x and NO_x simultaneously from flue gas after coal has been combusted, and to do the enhancements that we've identified in the other technologies.

Mr. NELSON. Can you tell me what portion of DOE's fossil energy program is devoted to R&D of retrofit control technologies as opposed to R&D of new control technologies?

Mr. MARES. Many of our technologies can be used in a retrofitable mode, and I do not remember that we have to date made an analysis of how you would split the amount of money between one versus the other. I would prefer, if I may, to go back and ask that, and then answer that for the record, sir.

[The information follows:]

RETROFIT VERSUS NEW CONTROL TECHNOLOGY

The Department of Energy's FY 1984 Fossil Energy program does not make the distinction in its R&D funding between retrofit control technology development and new control technology development. The R&D activities are focused toward the development of new technologies. However, many of these technologies could be used in a retrofit application. They include the following:

- Advanced Coal Cleaning - There are a number of fine coal cleaning techniques which have the potential for efficiently removing 95 percent of the ash and, hence, pyritic sulfur from coal. For high sulfur coal, this means that 50 to 70 percent of total sulfur can be removed. While the cleaned coal represents a higher cost product than the mother coal, this cost will be partly offset by improved plant availability, lower maintenance costs and lower transportation costs.

- Advanced Flue Gas Cleanup - DOE is developing a number of techniques for simultaneously removing 90 percent of the sulfur and 90 percent of the NO_x at a lower cost than commercially available options (lime/limestone scrubber plus selective catalytic reduction) while minimizing solid waste production by recycling the sorbent. Several options under development may be good candidates for retrofit in cases without severe space limitations.

- Gasification/Combined Cycle - This concept has the potential for being integrated with an existing power plant, if the existing boiler is replaced; sulfur and NO_x emissions are reduced, and the electrical output of the plant is increased.

- Atmospheric Fluidized Bed Combustion (AFBC) - This technology can be used to replace an existing boiler and extend the life of the plant while reducing sulfur emissions by 90 to 95 percent and NO_x emission by 50 to 75 percent. AFBC will not add to the operating/maintenance costs of the plant.

- Combustion Process Modification - DOE is developing several advanced combustors with control potential. Availability is expected within five years and has the potential for retrofit to at least part of the population of existing boilers.

- Pressurized Fluidized Bed/Combined Cycle - This concept has the potential for being integrated with an existing power plant, if the existing boiler is replaced; sulfur emissions reduced by 90 to 98 percent and NO_x emissions by 75 to 85 percent; and the electrical output of the plant is increased.

Mr. NELSON. Thank you, Mr. Chairman.

Mr. FUQUA. Thank you, Mr. Nelson.

Dr. Princiotta, thank you, and, Jan, we thank you for being here again.

The hearing is adjourned.

[Whereupon, at 12:24 p.m., the subcommittees recessed, to reconvene at the call of the Chairs.]

APPENDIX I

QUESTIONS AND ANSWERS SUBMITTED FOR THE RECORD

DON FUQUA (Fla.), Chairman

ROBERT A. ROE, N.J.
GEORGE E. BROWN, JR., CALIF.
JAMES H. SCHEUER, N.Y.
RICHARD L. OTTINGER, N.Y.
TOM HARKIN, IOWA
MARILYN LLOYD, TENN.
DOUG WALGREN, PA.
DAN GLICKMAN, KANS.
ALBERT GORE, JR., TENN.
ROBERT A. YOUNG, MO.
HAROLD L. VOLKMER, MO.
BILL NELSON, FLA.
STANLEY N. LUNDINE, N.Y.
RALPH M. HALL, TEX.
DAVE MC CURDY, OKLA.
MERVYN M. DYMALLY, CALIF.
PAUL SIMON, ILL.
NORMAN Y. MINETA, CALIF.
RICHARD J. DURBIN, ILL.
MICHAEL A. ANDREWS, TEX.
BUDDY MAC KAY, FLA.
T. ALTON VALENTINE, N.C.
HARRY M. REID, NEV.
ROBERT G. TORRICELLI, N.J.
FREDERICK C. BOUCHER, VA.

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September 29, 1983

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MANUEL LUJAN, JR., N. MEX.
ROBERT S. WALKER, PA.
WILLIAM CARNEY, N.Y.
F. JAMES SENSENBRENNER, WIS.
JUDD GREGG, N.H.
RAYMOND J. MC GRATH, N.Y.
JOE SKEEN, N. MEX.
CLAUDINE SCHNEIDER, R.I.
BILL LOWERY, CALIF.
ROD CHANDLER, WASH.
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J. M. POORE
Executive Director
ROBERT C. KETCHAM
General Counsel
DAVID S. JEFFERY
Minority Staff Director

Mohamed T. El-Ashry, Director
Environmental Quality Division
Tennessee Valley Authority
Knoxville, Tennessee 37902

Dear Dr. El-Ashry:

We want to thank you and Dr. High for your excellent testimony before the Subcommittee on Energy Development and Applications and the Subcommittee on Natural Resources, Agriculture Research, and Environment on September 20, on the subject of acid rain and its implications for fossil energy research and development policy. Attached are some questions that we would like you to answer, in order to supplement the record of the hearing on this topic.

We appreciate your cooperation on this matter. If you have any questions regarding this request, please contact Dr. Harlan Watson of the staff of the Subcommittee on Energy Development and Applications at (202) 225-4320, or Dr. Bob Palmer of the staff of the Subcommittee on Natural Resources, Agriculture Research, and Environment at (202) 226-6985.

Sincerely,

DON FUQUA, Chairman
Subcommittee on Energy
Development & Applications

JAMES H. SCHEUER, Chairman
Subcommittee on Natural Resources,
Agriculture Research & Environment

Questions for Dr. El-Ashry

1. In your testimony, you state that TVA has reduced the annual sulfur dioxide (SO_2) emissions from its coal-fired generating plants by about one million tons since 1978 at a capital cost of $724 million with an annualized cost of $334 million in order to comply with the Clean Air Act. Yet you also state that you have not been able to detect any trend of increasing or decreasing acidity, and that you have no direct evidence of a link between acid deposition and impacts to the resources in the Tennessee Valley.

   How then can you justify your call for further reductions in SO_2 and nitrogen oxide (NO_x) emissions at an additional substantial cost?

2. How much, if any, of the reduction in TVA's level of SO_2 emissions since 1978 was due to the retirement of old coal-fired plants?

3. Given the lack of knowledge about source/receptor relationships, what guarantee do you have that your emission reduction allocation scheme will actually result in a reduction in acidic deposition in the desired regions?

4. What is the schedule for construction and operation of the proposed AFBC demonstration facility at Shawnee?

5. Your preliminary cost estimate for construction of the AFBC demonstration facility at Shawnee is $200 million. What specific funding contributions to the facility do you expect from each of TVA, EPRI, the Commonwealth of Kentucky, and Duke Power?

   Do you expect funding from DOE, and if so, at what level?

6. The Senate version of the 1984 Interior Appropriations Bill includes $30 million for this AFBC demonstration facility. Are these funds really needed to construct the facility?

   What is the impact on the project if this $30 million is not obtained?

1. Question: In your testimony, you state that TVA has reduced the annual sulfur dioxide (SO_2) emissions from its coal-fired generating plants by about one million tons since 1978 at a capital cost of $724 million with an annualized cost of $334 million in order to comply with the Clean Air Act. Yet you also state that you have not been able to detect any trend of increasing or decreasing acidity, and that you have no direct evidence of a link between acid deposition and impacts to the resources in the Tennessee Valley.

   How then can you justify your call for further reductions in SO_2 and nitrogen oxide (NO_x) emmisions at an additional substantial cost?

   Answer: The 5-year period over which the million-ton reduction has been achieved is probably insufficient time to be able to discern a trend--particularly since this reduction occurred gradually and was not fully implemented until this year. However, we have been able to identify a reduction in gaseous sulfur (SO_2) deposition near our power plants where large emission reductions occured--indicating that dry deposition rates have decreased as a direct result of these emission reductions. A longer monitoring period will be needed to factor out the annual variations in meteorology from regional trends in rainfall acidity--particularly since two of the five years since 1978 were unusually dry.

   Moreover, substantial reductions from other sources contributing to the sulfur load in the Tennessee Valley region may be necessary before a statistically valid change in the trend of deposition is discernable. Analyses of TVA field studies indicate that the contribution of sulfur compounds from regions outside the TVA area play a major role in the observed atomspheric sulfur content in our region.

   Although we have no direct evidence of a link between acid deposition and impacts in the Tennessee Valley, circumstantial evidence available for acid sensitive areas in the eastern United States indicated possible irreversible damage to some of the Nation's most important natural resources and demonstrates a need for reduced acid deposition.

2. Question: How much, if any, of the reduction in TVA's level of SO_2 emissions since 1978 was due to the retirement of old coal-fired plants?

   Answer: None of the SO_2 emissions reduction referred to is due to retirement of coal-fired units. A combination of reduced generation by our coal-fired plants and increased generation by our nuclear units has, however, reduced the amount of generation required from our fossil plants and

further reduced our emissions by more than 50 percent. In fiscal year 1982 fossil-fired generation was 83 percent of the level in FY 1977 and SO_2 emissions were 49 percent of the FY 1977 level. Approximately one-third of the SO_2 emission reduction was due to reduced generation. The remaining two-thirds was a result of a combination of improved coal quality, heat rate improvement (i.e., more efficient use of coal), and installation of scrubbers.

3. Question: Given the lack of knowledge about source/receptor relationships, what guarantee do you have that your emission reduction allocation scheme will actually result in a reduction in acidic deposition in the desired regions?

   Answer: We have no guarantee that the emission reductions we believe warranted will result in reduced acid deposition in the desired region. In fact, we question whether any of the prediction techniques liekly to be available in the near future will be able to accurately quantify how much a given emission reduction will affect a given receptor area. However, given the evidence of a relationship between SO_2 and NO_x emissions and acidic deposition rates and the location of the acid sensitive regions with respect to the major emission sources, we feel reasonably certain that a reduction in these emissions will lead to reduced acid deposition in the desired region.

4. Question: What is the schedule for construction and operation of the proposed AFBC demonstration facility at Shawnee?

   Answer: Initial site preparation will start in April 1985 with steam to turbine in April 1989. A 3- to 4-year test program will commence in the latter part of 1989.

5. Question: Your preliminary cost estimate for construction of the AFBC demonstration facility at Shawnee is $200 million. What specific funding contributions to the facility do you expect from each of TVA, EPRI, the Commonwealth of Kentucky and Duke Power?

   Answer: Present commitments to the demonstration project include: $40 million from TVA, $75 million from EPRI, $10 million from Kentucky and $10 million from Duke Power.

   Question: Do you expect funding from DOE, and if so, at what level?

   Answer: If Congress passes the Senate version of the 1984 Interior Appropriations Bill, which includes $30 million for the AFBC demonstration facility, it is our understanding that these funds ($15 million in FY 1985 and $15 million in FY 1986) would come from DOE.

6. Question: The Senate version of the 1984 Interior Appropriations Bill includes $30 million for this AFBC demonstration facility. Are these funds really needed to construct the facility?

   Answer: The funds are needed to construct the facility. The $30 million requested will enable the project to proceed on schedule.

   Question: What is the impact on the project if this $30 million is not obtained?

   Answer: Without the $30 million in Federal funds, there is a risk that the remaining required funds would not be raised in a timely manner and the project schedule would have to be extended.

DON FUQUA (Fla.), Chairman

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September 29, 1983

Frank Princiotta, Director
Industrial Environmental Research Lab
U.S. Environmental Protection Agency
Research Triangle Park, North Carolina 27711

Dear Dr. Princiotta:

We want to thank you for your excellent testimony before the Subcommittee on Energy Development and Applications and the Subcommittee on Natural Resources, Agriculture Research, and Environment on September 20, on the subject of acid rain and its implications for fossil energy research and development policy. Attached are some questions that we would like you to answer, in order to supplement the record of the hearing on this topic.

We appreciate your cooperation on this matter. If you have any questions regarding this request, please contact Dr. Harlan Watson of the staff of the Subcommittee on Energy Development and Applications at (202) 225-4320, or Dr. Bob Palmer of the staff of the Subcommittee on Natural Resources, Agriculture Research, and Environment at (202) 226-6985.

Sincerely,

DON FUQUA, Chairman
Subcommittee on Energy
Development & Applications

JAMES H. SCHEUER, Chairman
Subcommittee on Natural Resources,
Agriculture Research & Environment

Questions for Dr. Princiotta

1. How does EPA coordinate its research efforts with those of DOE?

2. On page 7 of your testimony you state that EPA has recently demonstrated the ability of organic acid additives to further improve performance and reliability over state-of-the-art limestone scrubbers.

   Do these additives pose any additional environmental or safety problems?

3. What savings in capital costs does the spray dry FGD system offer over conventional FGD systems?

4. What has been the level of EPA funding of control technology R&D for FY 1980-FY 1983, and what level is planned for FY 1984?

   Please provide a detailed breakdown and description of these activities.

UNITED STATES ENVIRONMENTAL PROTECTION AGENCY
WASHINGTON, D.C. 20460

OCT 27 1983

OFFICE OF
RESEARCH AND DEVELOPMENT

Honorable Don Fuqua
Chairman, Subcommittee on Energy
Development & Applications
Committee on Science and Technology
House of Representatives
Washington, D.C. 20515

Dear Mr. Chairman:

Thank you for your letter dated September 29, 1983 to Mr. Frank Princiotta, Director, Industrial Environmental Research Laboratory Research Triangle Park, N.C. requesting additional information in order to supplement the record of the hearing held on September 20, 1983 before your committee. As you requested, we have prepared responses to your four questions which we are enclosing.

Mr. Princiotta appreciated the opportunity to participate in the joint subcommittee hearing on "Acid Rain and Its Implications for Fossil Energy Research and Development Policy." Please let us know if we can be of further assistance.

Sincerely yours,

David G. Stephan

for Courtney Riordan
Acting Assistant Administrator
for Research and Development

Enclosure

Question: 1. How does EPA coordinate its research efforts with those of DOE?

Answer: In 1978, OMB determined that the responsibility for energy related control technology development should be divided between DOE and EPA, with DOE assuming primary responsibility for any new commercial-scale hardware projects, while EPA would conduct development efforts only through the pilot-scale. This determination required DOE and EPA to develop a coordinated program to assure continued progress in development of these technologies. DOE/EPA cooperative efforts were formally initiated with an interagency agreement in 1979.

A cooperative plan (issued 10/79, revised 11/80) specified that DOE's Office of Coal Utilization and EPA's Office of Environmental Engineering and Technology hold working sessions every 6 months to review cooperative efforts, identify new areas of cooperation establish cooperative plans, and recommend funding levels for the next 2 fiscal years and to revise, if necessary, the cooperative plan. Meetings of the DOE/EPA working group were held 11/79, 05/80, 01/81, 05/81, and 01/82. The program during this period included significant pass-through funding from DOE to EPA with emphasis on SO_2 and NO_x demonstration projects.

Due to a reorganization at DOE and a major shift in program activities at DOE, the meetings were temporarily suspended but were resumed in 03/83. The recent coordination activities have focused on information exchange and have not included transfer of funds.

In addition to the cooperation in the energy/control technology area, DOE and EPA are cooperating on the Interagency Task Force for Acid Precipitation. Specifically, DOE (David Beecy, Fossil Energy) chairs Task Group B "Man-Made Sources" in which EPA plays a major supporting role; similarly EPA (Frank Princiotta, IERL-RTP) chairs the Task Group H "Control Technologies" which coordinates activities of DOE, EPA, and TVA regarding control technology development. Through these activities, EPA and DOE are very well aware of each others activities.

Question: 2. On page 7 of your testimony you state that EPA has recently demonstrated the ability of organic acid additives to further improve performance and reliability over state-of-the-art limestone scrubbers.

Do these additives pose any additional environmental or safety problems?

Answer: No significant incremental environmental impact is anticipated when adipic or other organic dibasic acids are used as additives in limestone scrubbers. The low concentrations of the acids in limestone slurries minimize any possibility of secondary environmental impacts.

In order to determine if solid waste from adipic acid enhanced limestone scrubbers should be classified as a hazardous waste, sludge samples were analyzed according to the Extraction Procedure Toxicity Test as prescribed by regulations pursuant to the Resource Conservation and Recovery Act (RCRA). Results showed that elements are not significantly different between extracts from sludges with and without adipic acid and that they are all considerably lower than the concentration deemed hazardous.

In order to evaluate further the impact of adipic acid addition, chemical and biological tests were conducted on samples collected while operating with and without adipic acid addition. Samples were taken from the flue gas after the scrubber, the oxidation tank vent, the scrubber slurry, and the filter cake. The chemical tests included the standard Level 1 test series. The biological tests included: (1) the mutagenicity assay, (2) CHO clonal toxicity assay, (3) rabbit alveolar macrophage (RAM) cytotoxicity assay, (4) human cell cytotoxicity assay, (5) rodent toxicity, and (6) fresh water toxicity assay on algae, fish, and invertebrates. These tests indicated that there was no significant difference in toxicity or mutagenicity between the samples containing adipic acid and its degradation products and samples from conventional limestone systems. Confirmatory genetic toxicity assays were also conducted on the leachate from adipic acid enhanced and conventional limestone FGD systems. The tests included salmonella/microsome assay, saccaromyces can^r/his^+ dual assay, and salmonella uvrB repair assay. The results from these tests showed no genetic activity in the samples.

It should be noted that adipic acid is included on the hazardous substances list which was promulgated by EPA pursuant to the Clean Water Act. This list was developed primarily to identify substances, which if discharged into lakes, rivers, and streams in sufficient quantities, would be toxic to aquatic organisms. The regulations require notification of EPA in the event of a discharge of the substance in greater than the designated reportable quantity. The reportable quantity for a discharge of adipic acid is 2270 kg (5000 lb). The fact that adipic acid is one of about 300 chemicals appearing on the hazardous substances list does not restrict its use in the nylon industry or even as a food additive. Likewise, it does not affect the use of adipic acid as an additive to FGD systems.

Worker exposure to adipic acid should not be a problem. Dust inhalation can be controlled by a filter mask if necessary. The item most often associated with using adipic acid in FGD systems is the unpleasant odor which has been reported. The odor is probably a result of adipic acid's being degraded to valeric acid which has a very low threshold for odor detection. The odor has been reported occasionally at Shawnee prototype tests and Springfield utility FGD demonstration; however, it has not been a problem at either location. The odor was not noticed in the testing at Rickenbacker industrial FGD demonstration. The odor, or possibility thereof, should not constrain use of adipic acid in FGD systems.

In summary, an extensive analytical program has verified that adipic or other organic dibasic acids do not pose any additional environmental or safety problems when used properly in FGD systems. In fact, the technology can reduce the volume of solid waste generated which reduces secondary environmental impacts. The technology has also shown that both capital and operating costs of limestone scrubbers can be reduced by 10% while increasing reliability and reducing energy requirements. This can translate to billions of dollars of savings to the utility industry and consumers.

Question: 3. What savings in capital costs does the spray dry FGD system offer over conventional (wet) FGD systems?

Answer: A 1981 study for EPA projected the capital costs of lime-based spray dryer FGD systems to be 15 to 25 percent lower than conventional wet limestone FGD systems for coals with sulfur contents of 0.7 to 3.5 percent. Over the same coal sulfur content range, the lime-based spray dryer systems were estimated to have annualized revenue requirements ranging from 25 percent lower for the low sulfur (0.7 and 0.9% S) coals to 5 percent higher for the 3.5 percent sulfur eastern coal. These costs are currently being updated; the updated costs, incorporating to the extent feasible actual costs from operational systems, are slated to be available early in 1984. We expect these estimates to show somewhat narrowed capital cost differentials as actual, rather than estimated, spray dryer costs become available. Annual revenue projections are expected to drop somewhat, however, since experience from the few full-scale spray dryers now operational indicate lower absorbent requirements than formerly estimated. These factors may extend the estimated break-even costs of dry and wet FGD systems to coals of sulfur contents greater than the 2 to 3 percent projected in the earlier studies. Since the spray dryer systems are less complex than conventional wet systems, they are also expected to be more reliable, but sufficient data to firmly substantiate this premise are not yet available.

Question: 4. What has been the level of EPA funding of control technology R&D for FY 1980 - FY 1983, and what level is planned for FY 1984?

Please provide a detailed breakdown and description of these activities.

Answer:

Level of EPA R&D Funding of Control Technology

Area	FY: 80	81	82	83	Planned 84
Multimedia	17.27	15.28	9.24	6.59	9.58
Oxidants	11.10	6.63	11.85	0.70	1.09
Hazardous Air Pollutants	3.58	2.22	0	0	0.20
Gases & Particles	7.63	6.72	7.55	3.25	2.42
Industrial Wastewater	2.78	1.43	0.76	0	0
Solid Waste (RCRA)	1.34	0.58	0	0	0
Municipal Spills	0.37	0.36	0.41	0	0

Description of the various areas are as follows:

Multimedia - Currently consists of two programs activities: (1) to develop and evaluate the Limestone Injection-through-a Multistage-Burner (LIMB) process. This technology should prove to be an inexpensive emission-control technology that will simultaneously remove sulfur oxides (SO_x) and nitrogen oxides (NO_x) from boiler flue gases of pulverized coal boilers; (2) to characterize and quantify the emissions and effluents from a variety of coal-based and oil-shale synthetic fuel processes and to identify and evaluate a variety of pollution-control technologies for these same processes.

Oxidants - A prefurnance combustion method for NO_x control with potential for SO_x control is being researched; and options of infurnace (post-flame) reburning for NO_x control on gas, oil and coal-fired, bench-scale combustors are being assessed for future potential application for field use.

Hazardous Air Pollutants - Research to evaluate the cost, performance and reliability of technologies for reducing or eliminating hazardous air-pollutant emissions from various industrial and combustion sources.

Gases and Particles - Research to define the basic mechanisms and principal parameters for two-stage electrostatic precipitator operations is being conducted. Research on fabric filtration is being conducted to define the mechanisms which make the Electrostatically Enhanced Fabric Filter (ESFF) work since filtration removal systems will be useful in combined SO_x/particle control. Pilot studies of flue-gas desulfurization (FGD) focus on the application of dry systems, and various adsorbents and additives to reduce the operational cost of spray drying and to increase its effectiveness for SO_x control.

Industrial Wastewater - The characterization of effluents from coal-fired steam-electric utility boilers and the development of the data base necessary for the establishment of Discharge Standards for these facilities.

Solid Waste (RCRA) - The assessment of the effectiveness control technology options for the disposal of fly ash and scrubber sludges to determine optimum requirements necessary to protect groundwater and surface water.

Municipal Spills - Research has focused on the development of techniques and procedures to prevent and contain hazardous releases.

DON FUQUA (Fla.), Chairman

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Minority Staff Director

September 29, 1983

Mr. Jan W. Mares
Assistant Secretary for Fossil Energy
U.S. Department of Energy
Washington, D.C. 20585

Dear Mr. Mares:

We want to thank you for your excellent testimony before the Subcommittee on Energy Development and Applications and the Subcommittee on Natural Resources, Agriculture Research, and Environment on September 20, on the subject of acid rain and its implications for fossil energy research and development policy. Attached are some questions that we would like you to answer, in order to supplement the record of the hearing on this topic.

We appreciate your cooperation on this matter. If you have any questions regarding this request, please contact Dr. Harlan Watson of the staff of the Subcommittee on Energy Development and Applications at (202) 225-4320, or Dr. Bob Palmer of the staff of the Subcommittee on Natural Resources, Agriculture Research, and Environment at (202) 226-6985.

Sincerely,

DON FUQUA, Chairman
Subcommittee on Energy
Development & Applications

JAMES H. SCHEUER, Chairman
Subcommittee on Natural Resources,
Agriculture Research & Environment

Questions for Mr. Jan Mares

1. You noted that the DOE's Office of Fossil Energy heads the Interagency Task Force on Acid Precipitation's man-made emissions task group.

   a. What are main research goals of this group?

   b. What key questions is this research addressing?

   c. What are this task group's short-term research objectives?

   d. What are this task group's long-term research objectives?

   e. What is this task group's research scope and strategy?

   f. What is the level of funding for this task group for each of the fiscal years 1982, 1983, and 1984? Please identify both the sponsoring agencies and the performers.

   g. What is the proposed implementation schedule of this task group's research pogram?

   h. What are the specific outputs of this task group's research?

2. What portion of DOE's Fossil Energy program is devoted to R&D of retrofit control technologies versus R&D of new control technologies?

3. On pages 5 and 6 of your testimony, you note that improvement of coal cleaning may require grinding to sizes finer than talcum powder. Aren't there significant environmental and safety hazards in dealing with such coal powders (e.g., coal dust explosions)?

4. In your testimony you note a number of pre-combustion and post-combustion cleaning technologies, as well as clean-up during combustion technologies being pursued by DOE. What is the relative maturity of each of these, and, on the basis on the state-of-the-science of each, how do they rank in terms of cost-effectiveness?

5. On page 15 of your testimony, you note that DOE "plans to commit nearly $3.25 million to the nearly $27.5 million of research to be conducted by the [Interagncy Task Force on Acid Precipitation] in FY 1984," in six task group areas.

   How much funding is provided for each of these areas by each of the three DOE offices (Office of Fossil Energy, Office of Energy Research, and Office of Environmental Protection, Safety, and Emergency Preparedness)?

   How does DOE decide which office pays for which portion of the research?

6. Is it DOE's intention to provide any funding for the proposed atmospheric fluidized bed combustion (AFBC) facility at TVA's Shawnee Steam Plant near Paducah, Kentucky, and if so, at what level?

7. On page 11 of your testimony you note that information is being collected from the 80-megawatt (thermal) pressurized fluidized bed (PFB) boiler in Grimethorpe, England. The House version of the FY 1984 Interior Appropriations Bill includes $2 million for additional testing. Is this a worthwhile expenditure or have we collected all the useful information we need from the project?

8. On page 11 of your testimony you mention that federal funds are being used to complete the construction of the 13-megawatt (electric) PFB pilot plant at Wood-Ridge, NJ, with the Curtiss-Wright Corporation having agreed to secure private funding for the plant's operations. We were under the impression that there was some delay in the project due to Curtiss-Wright's failure to come forward with their share of the funding. What is the status of the report?

9. How does DOE coordinate its research efforts in the area of control technology with those of EPA?

10. What has been the level of DOE's funding of control technology R&D for FY 1980-FY 1983, and what level is planned for FY 1984?

    Please provide a detailed breakdown and description of these activities.

HS&T Hearing of
Sept. 20, 1983
Mr. Mares Testified

Interagency Tack Force on Acid Precipitation

Question 1a: You noted that the DOE's Office of Fossil Energy heads the Interagency Task Force on Acid Precipitation's man-made emissions task group (8). What are main research goals of this group?

Answer: Task Group B will research man-made sources of species emitted into the atmosphere which are suspected to be participants in the formation of acid precipitation. Emission patterns of these species will be determined for the 48 contiguous states and for Canada. Patterns will be estimated for the past and for the present, and future patterns will be predicted for a number of bounding economic scenarios. A number of control scenarios will be evaluated in terms of their ability to alter future emission patterns and the cost of applying these controls will be estimated. A general description of the TG-B goals can be summarized as follows:

- o Provide, with adequate geographic, temporal and sectoral resolution, an accurate and complete inventory of emissions from man-made sources believed to be important in acid deposition processes; and,

- o Provide models which predict how these emissions may be altered by factors such as economic growth, fuel supply, emissions regulations, and control techniques, and which can calculate the cost of alternative control strategies.

Question 1b: What key questions is this research addressing?

Answer: Each research activity of Task Group B is directed to the resolution of a set of fundamental scientific questions, each of which must be answered in satisfactory detail before key policy questions can be resolved by the Interagency Task Force. Task Group B has developed the following list of key questions it must answer:

1. Which industries or activities are the major source of important pollutants?

2. Where are these major sources or source categories located?

3. What are the characteristics of each emission source or source category?

   a. Geographic differences in emission characteristics.

   b. Temporal differences in emission characteristics.

   c. Differences in operating characteristics between regions and during different time periods.

   d. Characteristics of major pollution control technologies.

4. How many tons of each pollutant are and will be emitted from each source in each geographic area during each time period?

5. How might the emissions from each major source or source category change in response to different government policies?

6. How much would it cost each source or source category to respond to such policies?

These questions provide a general guide to the results Task Group B is seeking from current and future research. Research designed to answer these questions, coupled with continuing dissemination, refinement and maintenance of the data collected, will meet the research goals of Task Group B.

Question 1c: What are this task group's short-term research objectives?

Answer: The Interagency Task Force is scheduled to perform major assessments of the acid deposition phenomenon in 1985, 1987 and 1989. Each of these assessments will employ the best data, models and scientific understanding available at the time, and will therefore yield progressively better answers.

The Task Group B program is coordinated with this schedule of assessments. In general, short-term research results will support the 1985 assessment (and meet other short-term needs). In the longer term, refinements of the emissions inventory and model set will contribute to a more detailed 1987 assessment. Further refinements will be accomplished prior to the 1989 assessment.

To support the 1985 assessment, Task Group B must develop an initial emissions inventory and emissions model set. The emissions inventory is also needed to test the Eulerian and Lagrangian models of atmospheric transport and transformation under development by Task Group C. Specific short-term research objectives include the following:

Emissions Inventory

1. In cooperation with other Task Groups and researchers, determine which pollutants must be included in the inventory, and what degree of geographic, temporal, and source category detail is needed. (This objective has been largely achieved.)

2. Identify the processes that are the principal sources of these pollutants, and determine the emission characteristics of these sources.

3. Inventory principal sources and calculate emissions in the 1980 base-year to the requisite degree of geographic, temporal, and source category detail.

4. Assemble the inventory of emissions into a readily accessible data base.

In summary, the short-term inventory research objective is to define and develop a 1980 base-year inventory at a level of resolution required for all purposes, including testing of the highly detailed Eulerian Model.

Emissions Model Set

5. Develop the test detailed models of the electric utility sector, industrial fuel combustion, and industrial process emissions of volatile organic compounds.

6. Develop and test simplified models of the residential and commercial combustion sectors and of the transportation sector.

7. Develop and test a simplified model to extend forecasts of emissions from all sources from the year 2000 to the year 2030.

8. Develop a capability to forecast economic growth and energy demand in relevant industries to the year 2030.

In summary, the short-term modeling research objective is to develop a simplified capability to project emissions from all important industries to the year 2030, within a consistent economic and energy forecast framework, and incorporating the most detailed models of individual industrial sectors available by 1985.

Question 1d: What are this task group's long-term research objectives?

Answer: By 1987, the Task Force will be prepared to perform the first complete and detailed assessment of the acid deposition phenomenon. Task Group B research objectives after 1985 are tailored to supporting that 1987 assessment, to refine the emissions inventory and model set for the 1989 assessment.

Emissions Inventory

9. Update emission inventory to 1984 base year.

10. Add additional pollutants as appropriate: ammonia, hydrogen flouride, hydrochoric acid, and alkaline substances are among candidates. Provide volatile organic compounds (VOC) by reactivity class and meet further disaggregation requirements.

The achievement of these long-term emissions inventory objectives will provide the Task Force with an accurate and resolute pattern of 1984 emission for use by Task Group I for use in accomplishing its 1987 and 1989 assessment, and for use by Task Group C in conjunction with 1984-deposition data to verify the accuracy of the Eulerian model.

Emissions Model Set

11. Refine, test and operate emissions model set for 1987 assessment.

12. Refine model set for 1989 assessment.

Accomplishments of long-term research objectives by Task Group B will meet all essential requirements for emissions data and industry emissions models presently enunciated by the intended recipients of Task Group B outputs, and will support development of the 1987 and 1989 assessments.

Question 1e: What is this task group's research scope and strategy?

Answer: As previously noted, the scope of the Task Group B effort is to identify the past, present and future man-made emissions patterns of species suspected to be participants in acid precipitation formation over an area including the 48 states and Canada. The species presently being included are: SO_2, SO_4 (acid and non-acid), NO_x, NO_2, NO, Total particulates, Volatile Organic Compounds in seven Reactivity Classes, HCl, NH_3 and Alkaline Dust.

The strategy in developing both the emissions inventory and the emissions models is to create these new products by building on existing inventories and models wherever possible. A thorough review of each product is planned prior to the release for use by ITFAP. This review will include review by groups outside of the ITFAP.

Question 1f: What is the level of funding for this task group for each of the fiscal years 1982, 1983, and 1984? Please identify both the sponsoring agencies and the performers.

Answer: Efforts in support of the ITFAP by TG-B, which were funded specifically under the ITFAP budget, are as follows:

	Funding in Thousands of Dollars		
	FY 1982	FY 1983	FY 1984
DOE	$ 300	$ 300	$ 260
EPA	$ 921	$ 1,050	$ 1,090
TOTAL	$ 1,221	$ 1,350	$ 1,350

DOE Performers: FY 1982 - K. Rahn, National Bureau of Standards, PEDCO Environmental, Inc., MITRE

FY 1983 - K. Rahn, National Bureau of Standards, PEDCO Environmental, Inc., Argonne National Lab, MITRE, Brookhaven National Lab, Thompson Ramo Woolridge, Pittsburgh Energy Technology Center (DOE)

FY 1984 - Argonne National Lab, MITRE, Brookhaven National Lab, Thompson Ramo Woolridge, Pittsburgh Energy Technology Center (DOE)

EPA Performers: FY 1982 - University Research Group on Energy, E. H. Pechan Assoc.

FY 1983 - University Research Group on Energy, Thompson Ramo Woolridge, Energy and Environmental Analysis, Inc., GCA Corp., Engineering Sciences, Inc., E. H. Pechan Assoc., Environmental Protection Agency/Research Triangle Park, Pacific Environmental Science

FY 1984 - University Research Group on Energy, Thompson Ramo Woolridge, Energy and Environmental Analysis, Inc., Radian, GCA Corp., Engineering Sciences, Inc., E. H. Pechan Assoc., Environmental Protection Agency/Research Triangle Park

Question 1g: What is the proposed implementation schedule of this task group's research program?

Answer: See Attached Figure 1 and Figure 2.

Question 1h: What are the specific outputs of this task group's research?

Answer: The specific outputs of the TG-B efforts as can be seen on the schedules, Figure 1 and Figure 2 are:

Emissions Inventory, 1980
Emissions Inventory, 1984
Emissions Model Set, 1985
Emissions Model Set, 1987
Emissions Model Set, 1989

FIGURE 1
EMISSIONS DATA BASE SCHEDULE

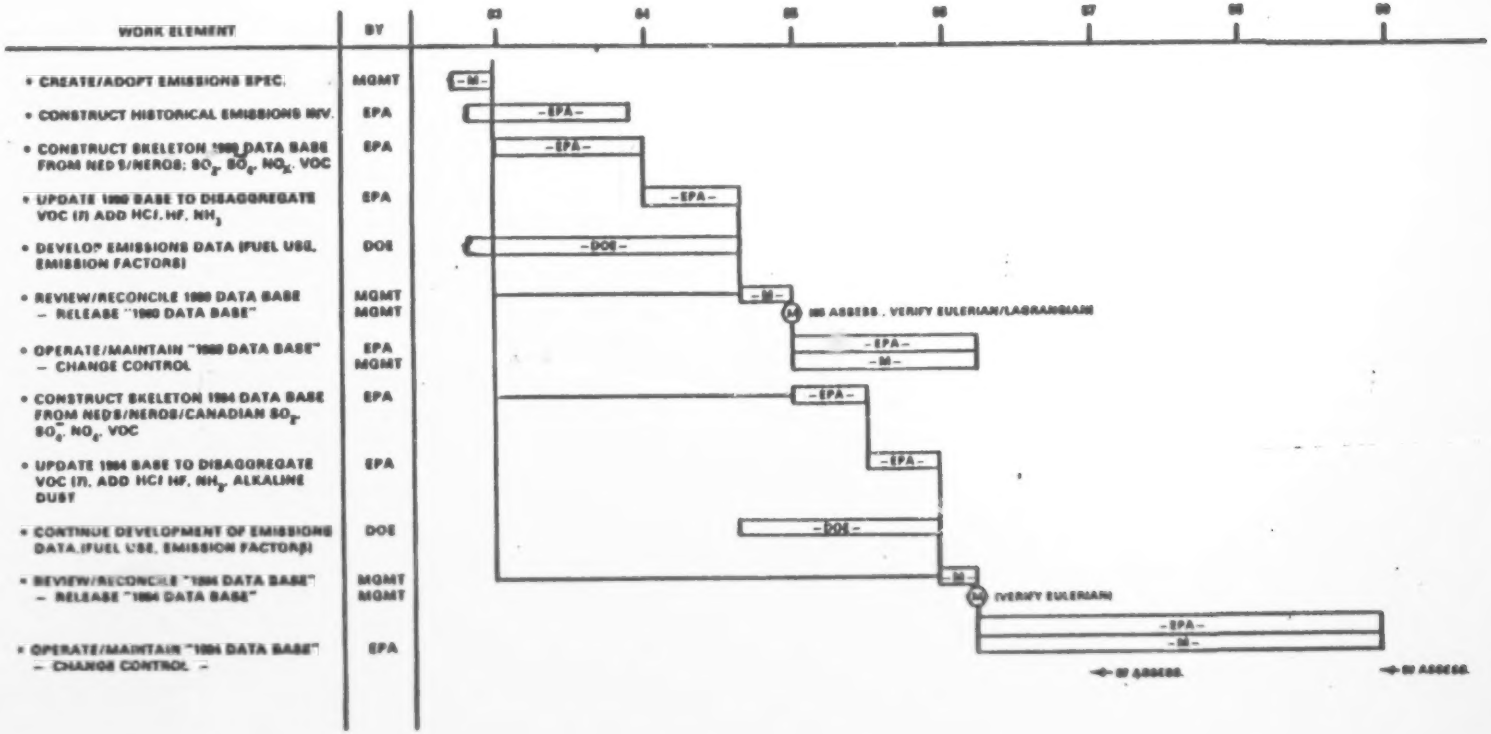
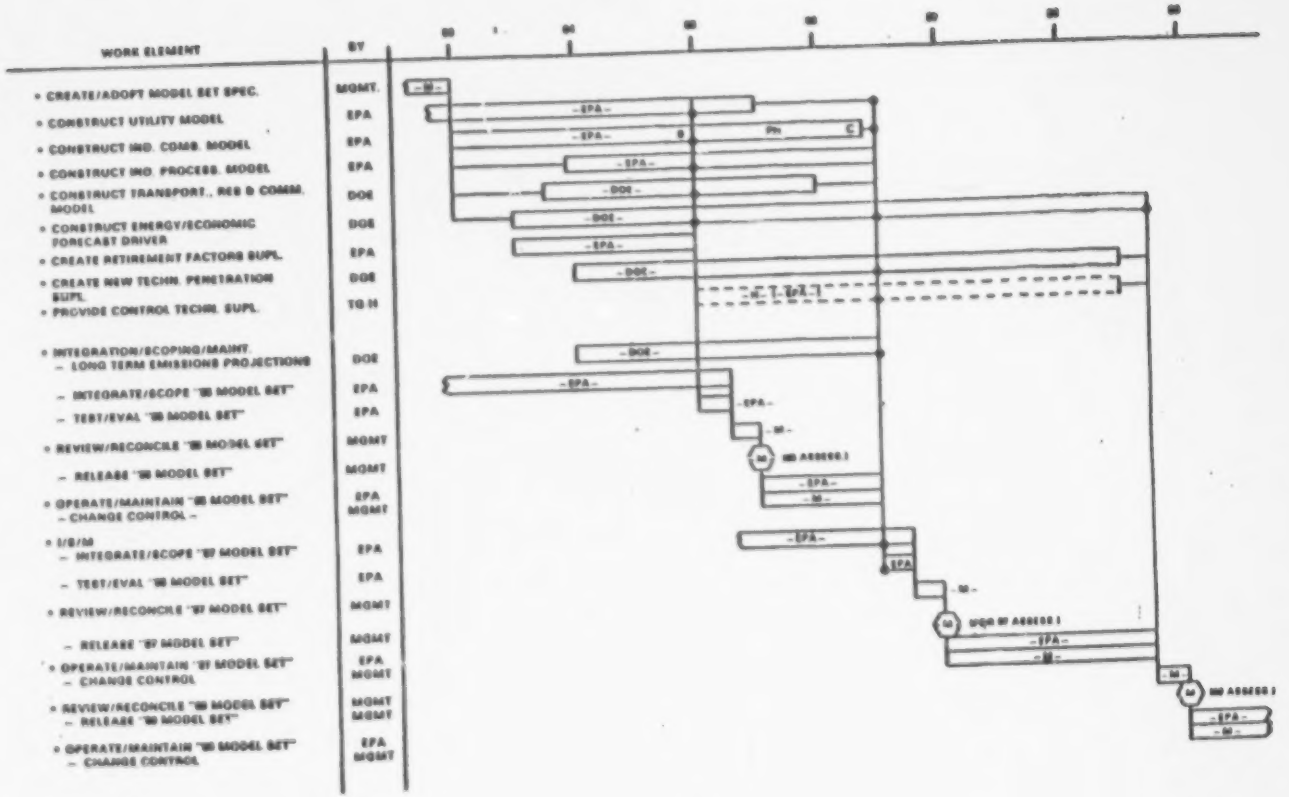


FIGURE 2
MODEL SET DEVELOPMENT SCHEDULE



RETROFIT VERSUS NEW CONTROL TECHNOLOGY

Question 2: What portion of DOE's Fossil Energy program is devoted to R&D of retrofit control technologies versus R&D of new control technologies?

Answer: The Department of Energy's FY 1984 Fossil Energy program does not make the distinction in its R&D funding between retrofit control technology development and new control technology development. The R&D activities are focused toward the development of new technologies. However, many of these technologies could be used in a retrofit application. They include the following:

- Advanced Coal Cleaning - There are a number of fine coal cleaning techniques which have the potential for efficiently removing 95 percent of the ash and, hence, pyritic sulfur from coal. For high sulfur coal, this means that 50 to 70 percent of total sulfur can be removed. While the cleaned coal represents a higher cost product than the mother coal, this cost will be partly offset by improved plant availability, lower maintenance costs and lower transportation costs.

- Advanced Flue Gas Cleanup - DOE is developing a number of techniques for simultaneously removing 90 percent of the sulfur and 90 percent of the NO_x at a lower cost than commercially available options (lime/limestone scrubber plus selective catalytic reduction) while minimizing solid waste production by recycling the sorbent. Several options under development may be good candidates for retrofit in cases without severe space limitations.

- Gasification/Combined Cycle - This concept has the potential for being integrated with an existing power plant, if the existing boiler is replaced; sulfur and NO_x emissions are reduced, and the electrical output of the plant is increased.

- Atmospheric Fluidized Bed Combustion (AFBC) - This technology can be used to replace an existing boiler and extend the life of the plant while reducing sulfur emissions by 90 to 95 percent and NO_x emission by 50 to 75 percent. AFBC will not add to the operating/maintenance costs of the plant.

- Combustion Process Modification - DOE is developing several advanced combustors with control potential. Availability is expected within five years and has the potential for retrofit to at least part of the population of existing boilers.

- Pressurized Fluidized Bed/Combined Cycle - This concept has the potential for being integrated with an existing power plant, if the existing boiler is replaced; sulfur emissions reduced by 90 to 98 percent and NO_x emissions by 75 to 85 percent; and the electrical output of the plant is increased.

Question 3: On pages 5 and 6 of your testimony, you note that improvement of coal cleaning may require grinding to sizes finer than talcum powder. Aren't there significant environmental and safety hazards in dealing with such coal powders (e.g., coal dust explosions)?

Answer: When coal is ground to sizes finer or as fine as talcum powder the environmental and safety hazards in dealing with such coal powders will be minimized by:

- operating in a closed-system
- operating under an inert atmosphere

These procedures involve state-of-the-art technology currently utilized by coal-fired electric utilities, i.e., the coal pulverizer/burner feed system.

Question 4: In your testimony you note a number of pre-combustion and post-combustion cleaning technologies, as well as cleanup during combustion technologies being pursued by DOE. What is the relative maturity of each of these, and, on the basis on the state-of-the-science of each, how do they rank in terms of cost-effectiveness?

Answer: The Department of Energy is funding several control technologies. Their estimated performance, cost and maturity is shown below:

Technology	Emissions Reduction (%) SOx	NOx	Incremental Cost (Mills/Kwhr)	Maturity
Limestone Scrubber	90-95	Neg.	13-20	Available
Dry Scrubber	70	Neg.	10-12	Available
Limestone Injected Modified Burner (LIMB)[1]	30-60	40-50	1-5	Commercial Early 90's
Advanced NO_x/SO_x Cleanup Devices	90-95	90-95	6-10	Commercial Early 90's
Conventional Physical Coal Cleaning	33-50	Neg.	2-5	Available
Advanced Physical Coal Cleaning	60-65	Neg.	4-7	Commercial Late 80's
Physical and Chemical Coal Cleaning	90-94	Neg.	13-20	Commercial Early 90's
Pressurized Fluidized Bed Combustion Retrofit	90-98	75-85	10[2]	Commercial Early 90's
Atmospheric Fluidized Bed Combustion Retrofit	90-95	50-75	12	Commercial Early 90's
Gasifier Combined Cycle/Gas Turbine Retrofit[3]	90-98+	Neg.[4]	15-25	Commercial Early 90's

1/ EPA funded.
2/ Does not include the benefits of lower cost additional capacity.
3/ Results in capacity expansion.
4/ NO_x reduction up to 70% may be obtainable with water injection.

Question 5: On page 15 of your testimony, you note that DOE "plans to commit nearly $3.25 million to the nearly $27.5 million of research to be conducted by the [Interagency Task Force on Acid Precipitation] in FY 1984," in six task group areas.

How much funding is provided for each of these areas by each of the three DOE offices (Office of Fossil Energy, Office of Energy Research, and Office of Environmental Protection, Safety, and Emergency Preparedness)?

How does DOE decide which office pays for which portion of the research?

Answer:

Funding ($ in thousands)

ITFAP Task Group	DOE Sponsoring Organization	FY 1984 Request
A. Natural Sources	None	$ 0
B. Man-Made Sources	Fossil Energy (FE)	260
C. Atmospheric Processes	Energy Research (ER)	610
D. Deposition Monitorirg	ER	602
E. Aquatic Impacts	None	550
F. Terrestrial Impacts	ER	50
G. Materials Research	None	0
H. Control Technologies	Non Applicable	0
I. Assessment/Policy Integration	ER	500
	Emergency Preparedness (EP)	675
		$3,247

Participation

Participation by offices within DOE is based on how closely the ITFAP need is associated with work normally performed in that office. Most ITFAP needs are met by a combination of new activities and extensions of current activities within research areas in which that office is already active for reasons other than acid precipitation concerns. For example, DOE fuel use pattern data can be extended by the use of emission factors to provide emissions data.

Combustion Systems

Question 6: Is it DOE's intention to provide any funding for the proposed atmospheric fluidized bed combustion (AFBC) facility at TVA's Shawnee Steam Plant near Paducah, Kentucky, and if so, at what level?

Answer: DOE is aware of the conference action on the FY 1984 Interior and Related Agencies Appropriations Bill (H.R. 3363) which "provides $15,000,000 in each of Fiscal Years 1985 and 1986 for a demonstration facility near Paducah, Kentucky, for the generation of electricity from coal using an atmospheric fluidized-bed combustion process..." DOE is developing contingency plans to incorporate the funds in this project and will coordinate with EPRI and TVA so that when the Fossil Energy Appropriations Bill has been signed into law, the necessary actions will be taken to insure that Congressional intent is carried out in regard to this program.

Combustion Systems

Question 7: On page 11 of your testimony you note that information is being collected from the 30-megawatt (thermal) pressurized fluidized bed (PFB) boiler in Grimethorpe, England. The House version of the FY 1984 Interior Appropriations Bill includes $2 million for additional testing. Is this a worthwhile expenditure or have we collected all the useful information we need from the project?

Answer: The present program (60 million pounds sterling funded equally by the U.S., U.K., and FRG) is scheduled to complete testing by May 1984 with final reports in November 1984. The Congressional Record of September 30, in reporting on the agreements of the conferees on the Department of Energy Fiscal Year 1984 appropriation, notes that $1,500,000 is provided for testing at Grimethorpe. When the Fossil Energy Appropriation Bill is signed into law, the necessary actions will be taken to insure that the Department works actively with Great Britian and West Germany to determine if additional testing at the Grimethorpe facility is worthwhile to improve the pressurized fluidized bed technology. Until these discussions occur, we do not believe a judgment can be made as to the usefulness of additional tests at the Grimethorpe faciiity.

Combustion Systems

Question 8: On page 11 of your testimony you mention that federal funds are being used to complete the construction of the 13-megawatt (electric) PFB pilot plant at Woodridge, N.J., with the Curtiss-Wright Corporation having agreed to secure private funding for the plant's operations. We were under the impression that there was some delay in the project due to Curtiss-Wright's failure to come forward with their share of the funding. What is the status of the report?

Answer: Curtiss-Wright has notified DOE regarding the closing of their facility in Woodridge, New Jersey; however, this would not and is not interferring with completion of construction which is on schedule. (A separate structure from the above mentioned Curtiss-Wright facility.)

DOE provided the funds to complete construction of the pilot plant. Curtiss-Wright, in their cooperative agreement with DOE, is required to make best efforts to obtain private sector funds for the operational phase of the pilot plant. DOE has heard unofficially that Curtiss-Wright is having difficulty in raising the funds to operate the pilot plant; however, DOE has not been officially notified by Curtiss-Wright as to their inability to operate the pilot plant.

Control Technology

Question 9: How does DOE coordinate its research efforts in the area of control technology with those of EPA?

Answer: Coordination of the two programs is formally accomplished in a number of ways. Joint program review and coordination meetings are held 1-2 times per year. Joint projects are cosponsored under an interagency agreement which was renewed last month (September 1983) for a four-year period. DOE has a member on the EPA-chaired Task Group H (Control Technology) of the Interagency Task Force for Acid Precipitation. DOE serves on the Industry Review Panel for the EPA Limestone Injected Multi-Stage Burner (LIMB) project. In addition, informal coordination is effected in a number of ways. Interchange of research and development reports is conducted on a routine basis, as well as assistance in the review and evaluation of proposals.

Control Technology

Question 10: What has been the level of DOE's funding of control technology R&D for FY 1980-FY 1983, and what level is planned for FY 1984?

Please provide a detailed breakdown and description of these activities.

Answer: Funding for Control Technology research and development in the area of acid rain precipitation is as follows:

($ in thousands)

	FY 1980	FY 1981	FY 1982	FY 1983	FY 1984
(Flue Gas Cleanup)	$25,050	$15,000	$ 6,240	$ 7,000	$ 8,100

A detailed description of FY 1984 work to be done in the area of acid rain is as follows:

FY 1984 Flue Gas Cleanup Program:

o Systems and Economic Analysis	$ 534
o Advanced NO_x/SO_x Removal Chemistry	660
o Flue Gas Cleanup for Western Coals	875
o Fluidized Bed Copper Oxide Process Development	700
o Electron Beam/Lime Spray Dryer Proof of Concept	589
o Evaluation of Microwave Enhancement for Flue Gas Cleanup Applications	162
o Electron Beam/Ammonia Proof of Concept	1,629
o Electron Beam Particle Charging	200
o Integrated Environmental Control Modeling and Assessment	220
o Thermodynamic/Thermochemical Properties of Compounds for Flue Gas Cleanup Processes	150
o Chelating Agents for Nitric Oxide Control	124
o NO_x/SO_x Particulate Systems Integration Study	300
o Evaluation of the Applicability of Electromagnetic Enhancement of Flue Gas Cleanup Processes	300
o Evaluation of Combined NO_x/SO_x Processes Based on the use of Sodium Compounds	1,157
o Comprenensive Review and Analysis of Studies on Acid Rain	500
	$7,600

APPENDIX II

ADDITIONAL MATERIALS SUBMITTED FOR THE RECORD

DEPARTMENT OF AGRICULTURE

NATIONAL OCEANIC AND ATMOSPHERIC ADMINISTRATION

ENVIRONMENTAL PROTECTION AGENCY

Department of the Interior
Department of Health and Human Services
Department of Commerce
Department of Energy
Department of State
National Aeronautics and Space Administration
Council on Environmental Quality
National Science Foundation
Tennessee Valley Authority

Annual Report, 1982

prepared by

Interagency Task Force on Acid Precipitation

Washington, D.C.

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Executive Summary

This statutory Annual Report to the President and the Congress by the Interagency Task Force on Acid Precipitation marks the end of the first full year of an integrated research effort by the Federal agencies. The National Acid Precipitation Assessment Program is coordinated by the Task Force and focuses federally-funded research on the timely development of a firmer scientific basis for policy decisions.

This report summarizes the major research findings and accomplishments of the National Program's first year, with particular emphasis on their bearing on acid precipitation policy questions. It provides a brief overview of the available scientific information relevant to policy formulation. As the National Program progresses, future annual reports will provide updates and increasingly better information to assist decisionmakers and the public in continuing to resolve the "acid rain" issue. The program will integrate findings from diverse research areas in order to arrive at successively better answers to key questions, such as:

- What are the current and potential effects of acid deposition within each region, and to what extent could these be ameliorated by lower deposition levels or other mitigation methods?
- What is the quantitative relationship between the release of certain pollutants into the atmosphere and the amount of acidic materials that are deposited?
- What are the costs and environmental impacts of these effects, as well as the costs and benefits of controlling pollutant releases?

In accordance with the Acid Precipitation Act of 1980 (P.L. 96-294), the Task Force is vigorously implementing a comprehensive research program. Federally-funded acid deposition research has doubled, from about $11 million in FY 1980 to $22.3 million for FY 1983. The National Program is producing results at an increasing rate, disseminating them as soon as they become available. However, as recognized in the Act, it will take a systematic effort over a number of years to adequately address all the major uncertainties about the causes, effects, and management of acid deposition.

STATE OF THE SCIENCE

This overview on the current state of the science is based on information from all available sources, including research supported by Federal agencies, States, environmental organizations, private sector groups, and other nations.

Causes

Man-made atmospheric pollutants are probably the major contributors to acid deposition in northeastern North America. The approximate coincidence of the region of highest precipitation acidity with the areas of greatest sulfur dioxide and nitrogen oxides emissions provides circumstantial evidence to support this assertion. This conclusion is also supported by preliminary meteorological studies tracking the physical movement of one pollutant (sulfur dioxide) from sources of emission to suspected areas of deposition.

The National Program is also studying another contributory pollutant (oxides of nitrogen) and chemical substances suspected of being factors in controlling acid deposition. Current data and available methods, however, are not sufficient to quantify relationships between pollutant emissions and acid deposition on a regional scale, or under varying conditions. Nor is it yet possible to identify the specific change in acid deposition patterns that would result from a given change in precursor emissions.

In addition to man-made sources, natural sources of acid precursors are known to exist. On a global scale, it is roughly estimated that half the sulfur in the atmosphere is from natural emissions. Regionally, preliminary findings suggest that natural sources might contribute significantly to the acidity of precipitation in some areas, such as the southeastern United States. In the northeastern United States natural sources are believed to be minor contributors to acid deposition. The National Program includes research to increase our meager knowledge of the strengths, character, and distribution of natural sources of sulfur and nitrogen that can contribute to acidity as well as alkaline dusts that neutralize acids in precipitation. This information will be used to assess the potential significance of these sources in contributing to acid deposition in sensitive areas.

Source/Receptor Relationships

To formulate scientifically based control strategies, the relationship between emission of precursor pollutants and deposition of acidic material at sensitive receptors should be reliably established.

The ability of current generation models to predict source/receptor relationships with adequate resolution and accuracy is an unresolved issue. A study on the ability of eight models to predict the deposition of sulfur emissions in eastern North America found only a few suitable monitoring data points. While a method is now available to properly evaluate model performance, more data (quality assured and available as a time series) is needed to quantify the uncertainties in model predictions. In addition there are widely differing views of the relative importance of local sources (acid deposition from nearby sources of pollution) and long-range transport (acid deposition from far-removed sources of pollution) on the receptors in the local region.

Because of the urgent need to improve our ability to make predictions that reasonably estimate the change in amounts of acid deposition expected from decreases in pollution levels, the National Program includes several simultaneous approaches:

1. For near-term assessments, modifying current-generation models to refine their accommodation of chemical and physical factors over shorter periods of time;
2. Constructing a new generation of models to more effectively treat the physical movement of pollutants in the atmosphere, as well as the mechanisms for their conversion to acids and deposition;
3. Designing a series of field experiments to provide empirical, quantitative evidence of the relationship of pollution source to acid deposition; and
4. Upgrading and evaluating a suitable local/mesoscale model to quantify the relative contribution of local versus long-range transport on receptors.

Intensive efforts underway in the National Program and elsewhere should enhance our ability to project the consequences of changes in emission patterns.

Impacts

Among the possible effects of acid deposition, aquatic impacts are currently of greatest concern because of the sensitivity of certain surface waters to acidification and the known potential for damage to fish and other organisms. Initial surveys indicate that a small number of lakes concentrated in the sensitive regions of northeastern North America have been acidified, and their biota have changed.

In the Adirondacks, one of the most sensitive regions in North America, the State of New York has classified 3.6 percent of lake area as in "critical" condition (7,466 acres of 246,388 acres total) and 14 percent endangered; 84 percent were surveyed. Acid deposition is probably the major contributor to this acidification, but the rates of change and the factors controlling it are still poorly understood.

Many of the reported cases that suggest acid deposition is the cause of some observed change in an aquatic ecosystem are based on circumstantial evidence and lack documentation of the mechanisms linking cause and effect. At present, the extent of actual aquatic damages is not well established. The question of long-term trends in freshwater acidification also remains uncertain.

The National Program produced maps in FY 1982 depicting the regions where surface waters could be most susceptible to acidification. Studies underway will better define the factors that control acidification, and improved regional monitoring and testing programs will produce more detailed and reliable inventories of the extent of damages and the location and value of resources at risk.

Beyond the alteration of the chemistry and biology of certain sensitive surface waters, the other effects of acid deposition in North America are undetermined. There is concern about possible effects on crops, forests, wetlands, soils, building materials, and indirect effects on human health (through drinking water or food). Although mechanisms for such effects are known, evidence has not yet demonstrated that they are widely occurring under ambient acid deposition in North America. For instance, although experimental studies with simulated acid rain have shown both positive and negative effects on crop species, effects on crops growing under actual field conditions have not been determined.

Similarly, although a dieback of spruce trees on mountain sites in New England is observable, the relative contribution of potential causes—acid deposition, drought, disease, and other pollutants—is still undetermined. In contrast to acid deposition, the effects of ozone and other air pollutants on crops and forests are well documented. The National Program is speeding up investigations and analyses to determine the actual effects of acid deposition.

RESEARCH PROGRESS AND OUTLOOK

In addition to the preceding synopsis of the state of knowledge about acid deposition, the "Research Progress and Outlook" part of this report describes in more detail current findings in each subject area. It also outlines many of the major research accomplishments of the National Program.

National Program FY 1982 Accomplishments

Some selected highlights of progress in FY 1982 for each research area follow.

A. Natural Sources:

- Developed and tested instruments for reliable field measurement of sulfur and nitrogen emissions from natural sources.
- Obtained preliminary measurements of natural sulfur compounds in the ocean that suggest emissions from marine sources may contribute significantly to atmospheric acidity in some areas.

B. Man-made Sources:

- Completed preliminary study on emission sources that suggested that nearby petroleum combustion and local and distant coal combustion could significantly affect local acid deposition.

• Continued development of models to predict the cost and emissions changes from utilities and industries based on various possible control strategies.

• Developed specifications for emissions inventories with improved spatial, temporal, and source-type resolution, and initiated studies to better characterize the man-made sources of acid precursor pollutants.

C. Atmospheric Processes:

• Obtained initial observations that confirm the importance of clouds in transforming sulfur dioxide to acidic sulfates.

• Initiated field studies that suggest the importance of man-made aerosols in cloud formation.

• Completed planning and initial testing for a major field experiment to be performed in FY 1983 that will track the movement of tracer gases over hundreds of miles after their release in the Midwest.

• Improved atmospheric models and used them to gain a better understanding of the movement and conversion of pollutants and the deposition of acidic material.

D. Deposition Monitoring:

• Produced the first comprehensive maps describing the distribution of major chemical substances over North America.

• Completed design for the National Trends Network, which will be established in FY 1983 building on existing National Atmospheric Deposition Program (NADP) sites and others that meet specific criteria.

• Established four research sites to test improved methods of monitoring wet and dry deposition.

• Compared field measurements of dry deposition obtained from different prototype methods and continued efforts to develop reliable techniques for routine measurement of dry deposition.

E. Aquatic Effects:

• Produced a nationwide map indicating regions where surface waters are likely to be most sensitive to acidification.

• Began long-term monitoring of aquatic resources (chemistry and biota) in key sensitive regions.

• Completed preliminary survey of drinking water in the Northeast that indicates some reservoirs and their watersheds have limited ability to buffer acid deposition.

• Developed better understanding of factors controlling the susceptibility of surface waters to acidification and evaluated models for predicting acidification.

• Evaluated existing information on mitigation strategies (liming), and initiated field and laboratory studies to examine their effectiveness and limitations.

• Began studies of acid deposition effects in the southeastern United States.

F. Terrestrial Effects:

• Initiated extensive long-term studies to determine whether acid rain has caused changes in rate of growth and species composition in forests.

• Completed a study evaluating whether acid rain predisposes pine seedlings to succumb to stress factors. It did not show increased susceptibility to insect or disease attack; some seedlings showed increased resistance to disease.

• Conducted agricultural experiments using simulated acid rain that did not indicate injury to potato plants or two forage plants, but decreased the yield in soybeans and corn under some conditions and had no effect under others.

• Performed soil studies in the Southeast, mid-Atlantic coast, and Northeast that showed response to acid rain varied according to mobilization of toxic metals and leaching of nutrients.

• Conducted studies indicating that southeastern watersheds are accumulating sulfur. The importance of this change on the chemical and biological characteristics of surface waters is not clearly understood.

• Completed several studies showing that forest ecosystems alter the acidity of rain traveling through them.

G. Effects on Materials and Cultural Resources:

• Began work to determine possible damage to historical monuments that is related to acid deposition.

• Established several field sites for testing materials and estimating the relationship of deterioration rates to acid deposition.

• Began analyzing available materials deterioration data and documenting the performance of possible protective treatments for masonry.

• Tested a simplified deposition monitor for application to materials damage research.

H. Control Technologies: (FY 1982 developments outside the National Acid Precipitation Assessment Program under preexisting agency pollution control hardware programs relevant to acid deposition)

• Successfully tested developmental prototype of low-cost, retrofitable technology (LIMB) that can simultaneously decrease sulfur dioxide and nitrogen oxides emissions from utilities by 50 percent. (EPA)

• Developed burner systems to decrease nitrogen oxides emissions by 75 percent in large coal-fired boilers. (EPA)

• Assembled data on the characteristics of U.S. coals to aid in determining their emission control potential. (EPA, DOE)

• Evaluated improved wet and dry sulfur dioxide scrubbing (removal) processes. (DOE, TVA, EPA)

I. Assessments and Policy Analysis:

• Prepared the "Critical Assessment Document: The Acidic Deposition Phenomenon and Its Effects" requested by the Clean Air Science Advisory Committee prior to formation of the National Program. Several chapters review acid deposition and its impacts. Contributors include 54 scientists from many institutions in this country and abroad. The document is now undergoing public review and final revision.

• Initiated work on developing methodologies for conducting integrated assessments of the significance of various scientific and economic information.

• Developed the preliminary version of a model for assessing sulfur dioxide emissions from the utility sector.

• Began investigation of estimating uncertainty in models and the implications for formulating management strategies.

J. International Activities:

• Established basis and mechanisms for increased cooperation between the United States and Canada on research and monitoring activities.

• Began developing specific proposals for joint research efforts with other nations.

• Published *International Directory of Acid Rain Researchers,* listing 1,600 scientists in 32 countries.

This annual report contains more details of the accomplishments in the various research categories in Sections A through J of the "Research Progress and Outlook" chapter. The Task Force also has made significant progress in FY 1982 in integrating and focusing Federal activities and coordinating them more effectively with private sector, State, and international acid deposition research efforts. In FY 1982 the Task Force met with representatives of these key groups to increase coordination. These accomplishments are described in the "Organization and Management Activities" chapters.

Outlook

In the next several years the research under the National Program will produce a number of important results. From FY 1983 to FY 1985 the Task Force agencies expect to accomplish several goals:

• Produce a report estimating how effective control of sulfur oxides, nitrogen oxides, and/or volatile organic compounds would be in reducing acid deposition for different regions.

• Complete assessments of the potential significance of alkaline dusts and natural sulfur and nitrogen emissions in influencing precipitation acidity over sensitive regions.

• Begin applying advanced models to predict utility emissions and develop a preliminary model to estimate industrial combustion emissions.

• Complete a major transport experiment (CAPTEX) to help determine long-range pollutant transport patterns and facilitate evaluation of atmospheric models.

• Develop a model capable of estimating local/mesoscale atmospheric deposition.

• Complete the National Trends Network, to provide adequate coverage to produce reliable annual maps of acid deposition for North America.

• Develop and begin applying improved techniques for monitoring dry deposition.

• Complete inventories and detailed regional maps of the extent of sensitive and currently acidified surface water.

• Prepare a comprehensive report on the status of drinking water supplies.

• Finish a broad assessment of the potential effects of acid deposition on soils.

• Establish dose-response information on corn and soybeans in eastern and midwestern regions of the United States.

• Complete initial determination of the effect of acid deposition on the susceptibility of forests to drought and other stresses.

• Prepare a report on historical rates of deterioration of building material and complete inventory of materials at risk in the Northeast.

• Complete initial assessment of the scope of material damages caused by acid deposition.

• Finish report on historical trends in emissions, deposition, and effects.

• Develop and apply advanced integrated assessment methods to evaluate the optimal strategies for managing acid deposition and its effects.

These and the many other achievements of the National Program will significantly improve our understanding of acid deposition and help develop sound policies to address the problem.

The National Acid Precipitation Assessment Program made an ambitious start and broad progress during its first year. The groundwork has been laid for continued progress in answering the scientific questions associated with acid deposition. A framework has been established to organize and integrate research results to address policy questions. The National Program's recurring assessments will provide a successively refined scientific basis for decisionmaking as well as opportunities to reexamine research directions.

Organization and Management Activities

HISTORY

The Interagency Task Force on Acid Precipitation was established by the Acid Precipitation Act of 1980 (P.L. 96-294) to plan and manage the National Acid Precipitation Assessment Program. Planning for the National Program began in October 1980, when the Task Force was first organized. In accordance with the Act, the Task Force drafted a National Acid Precipitation Assessment Plan, outlining the proposed research effort. That plan underwent extensive public review and the final version was submitted to Congress in June 1982.

The Task Force developed its first interagency budget for the Program in January 1981 and submitted it to Congress as part of the President's FY 1982 budget request. In January 1982, the Task Force issued its first annual report, outlining initial progress in the organization and planning of the National Program. The National Program's integrated research effort began in October 1981 with the beginning of FY 1982.

TASK FORCE

The Task Force oversees the planning and implementation of the National Acid Precipitation Assessment Program. The 20 members include one high-level representative from each of the 12 Federal entities in the Program, the directors of Argonne, Brookhaven, Oak Ridge, and Pacific Northwest National Laboratories, and four Presidential appointees. The Task Force is jointly chaired by the National Oceanic and Atmospheric Administration (NOAA), the Department of Agriculture (DOA), and the Environmental Protection Agency (EPA). Other participating Federal entities include the Departments of Interior (DOI), Health and Human Services (HHS), Commerce (DOC), Energy (DOE), and State (DOS); the National Aeronautics and Space Administration (NASA); the Council on Environmental Quality (CEQ); the National Science Foundation (NSF); and the Tennessee Valley Authority (TVA).

The primary responsibilities of the Task Force are:

- Plan and manage the National Acid Precipitation Assessment Program.
- Provide annual reports on the National Program's progress.
- Produce an annual interagency budget for the National Program.
- Coordinate the National Program with the research and monitoring activities of the private sector groups, environmental organizations, States, and other nations.
- Maintain an inventory of federally-funded acid precipitation research projects.
- Disseminate research results and assessments of their implications.

The role of the Task Force in planning the interagency budget for the National Program is a highly effective and unique aspect of the Federal effort. By working together through the Task Force, the agencies have established a research program that addresses national needs while building on the research expertise of the individual agencies. This strong interagency planning process avoids unnecessary duplication and eliminates crucial omissions in the National Program.

The Task Force sets the research goals for the National Acid Precipitation Assessment Program, identifies the projects needed to meet these goals, and decides which agencies are best suited to conduct the necessary work. The result is a comprehensive program of interlocking projects, with each agency contributing to specific aspects of the overall national effort.

During FY 1982 the Task Force met six times to review and approve the final National Acid Precipitation Assessment Plan, develop and approve the FY 1984 budget request, review and approve plans for the First Annual Review Meeting, and begin developing a detailed technical Operating Research Plan for the National Program. Appendix II lists all major Task Force activities with their dates and purposes.

TECHNICAL TASK GROUPS

The Task Force has 10 working-level Task Groups, one for each of the National Program's nine research categories and one for international activities. These technical groups include program managers and experts from all the participating Federal agencies and National Laboratories. They are responsible for detailed planning

and work in their assigned areas. A Coordinating Agency serves as the contact point within each Task Group. The Task Groups and their Coordinating Agencies are:

Task Group	Coordinating Agency
A. Natural Sources	NOAA
B. Man-made Sources	DOE
C. Atmospheric Processes	NOAA
D. Deposition Monitoring	DOI
E. Aquatic Effects	EPA
F. Terrestrial Effects	DOA
G. Effects on Materials and Cultural Resources	DOI
H. Control Technologies	EPA
I. Assessments and Policy Analysis	EPA
J. International Activities	DOS

These Task Groups met numerous times throughout FY 1982 and provided the material used in developing this report.

RESEARCH COORDINATION COUNCIL

The Research Coordination Council (RCC) oversees and integrates the efforts of the various Task Groups and develops draft reports, program plans, budgets, and other recommendations for consideration by the full Task Force. The joint chairs designated the Task Force's executive director to oversee its efforts. The RCC includes the leaders of all the task groups, the chairperson of the National Laboratory Consortium, and other appropriate agency representatives.

The Research Coordination Council met 10 times in 1982 and tackled many different tasks, including:

- Revising the National Acid Precipitation Assessment Plan;
- Drafting the Operating Research Plan;
- Planning the First Annual Review Meeting;
- Designing an inventory of Federal and State acid deposition research;
- Developing the FY 1984 interagency budget request;
- Drafting the 1982 Annual Report to the President and Congress.

The RCC will address many important management issues during the coming year, including plans for intensifying program review activities.

PROGRAM COORDINATION OFFICE

The Program Coordination Office staffs the Task Force and coordinates interagency activities. The office also disseminates information on the Program and is the primary liaison with States, private groups, the public, and other countries. The office is housed at CEQ and is funded by NOAA, EPA, DOA, DOI, and DOE.

During FY 1982, the Program Coordination Office supervised planning for the Task Force's First Annual Review Meeting (held in September 1982), and worked with non-Federal groups in organizing the Acid Precipitation Research Coordination Workshop (held in November 1982). The Program Coordination Office actively distributed thousands of copies of Task Force documents and responded to hundreds of requests for information on the National Program.

NATIONAL LABORATORY CONSORTIUM

The four National Laboratories on the Task Force form the Consortium required by law to integrate their acid precipitation research efforts and assist the Task Force in planning and conducting research and assessment activities. Each year the Consortium elects a chairperson to serve on the Research Coordination Council.

During FY 1982 members of the National Laboratories participated in the activities of all the Task Groups and assisted the Task Force in drafting the Operating Research Plan. The National Laboratory Consortium is studying data management needs for the National Program and inventorying Federal and State acid rain research.

MAJOR FY 1982 PROGRAM DEVELOPMENT ACTIVITIES

In addition to regular meeting and planning activities, the Task Force has undertaken several major efforts to improve the management and coordination of the National Program, including:

1. **Improving Coordination with Non-Federal Activities.** The Task Force is committed to conducting a National Program that coordinates Federal efforts with the research and monitoring activities of State and local governments, the private sector, environmental organizations, and other countries. Several specific steps have been taken to develop and encourage such cooperation and more extensive joint planning activities are anticipated. Besides reviewing the draft National Plan, the Task Force's April 1981 workshop of non-Federal experts began a dialogue with State and private groups. The workshop report describes the participants' ideas concerning how coordination and cooperation can best be accomplished. The Task Force is implementing many of these suggestions and is actively pursuing continued exchanges of information with non-Federal groups.

At the First Annual Review Meeting of the Task Force (September 1982), approximately 30 non-Federal participants provided valuable ideas to discussions of interrelationships among research areas, using the draft Operating Research Plan as background. Substantial research on acid deposition is sponsored by the private sector, certain States, and Canada; Task Force representatives have met with these non-Federal groups. These meetings have focused on reviewing programs and establishing a basis for better understanding and closer coordination. During the last year, the two most significant such events were.

- *Task Force-Canadian Research Program Cooperation.* In April 1982, Task Force representatives met with the Canadian Federal/Provincial Research and Monitor-

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ing Coordinating Committee (RMCC) to discuss bilateral scientific cooperation. Leaders of both the U.S. and Canadian research efforts emphasized the mutual value of greater cooperation and coordination of activities.

• *Task Force-State-Private Sector Coordination.* The Task Force initiated and encouraged joint sponsorship of an Acid Deposition Research Coordination Workshop in November 1982. The non-Federal sponsors included the Electric Power Research Institute, the American Petroleum Institute, the Gas Research Institute, the Coordinating Research Council, the National Council of the Paper Industry for Air and Stream Improvement, and the Keystone Center. The States, private sector, and the Federal Task Force were represented about equally. Environmental groups were also invited to participate.

The purpose of the workshop was to bring together key Federal and non-Federal managers of acid deposition research to review and discuss the management and coordination of their research planning efforts. Specifically, the two goals of the meeting were (1) to assess the scope of the nationwide acid deposition research effort, and (2) to initiate a continuing process for joint planning and coordination of Federal and non-Federal acid deposition research efforts.

In addition to developing joint statements on research needs and disseminating information, the State representatives agreed to organize by regions and to define their specific areas of expertise. The Task Force agreed to investigate ways to increase opportunities for State and private sector participation in the National Program planning and review groups. Followup activities include further discussion between the Task Force and the private sector and State representatives to work out the details.

2. **Developing a Detailed Operating Research Plan.** To ensure that diverse scientific findings are systematically organized to address the relevant policy questions, the Task Force is developing an Operating Research Plan. This detailed document supplements the more general National Acid Precipitation Assessment Plan (June 1982) and describes each project and how it relates to other activities and specific program outputs. This plan gives the technical and programmatic details on how the National Program's objectives will be met. The completed plan will be a dynamic document that will evolve with the Program and be updated annually to track the National Program and project plans for future years.

3. **Developing an Inventory of All Federal and State-Sponsored Acid Deposition Research.** The initial tabulation was completed this spring and the inventory will be maintained and updated for use by the Task Force and all interested parties. This inventory includes information on all ongoing projects with the performer(s), duration, funding, study area, research goals, methodologies, and expected outputs. The private sector has conducted a similar inventory of its activities; together with the Task Force's effort, this should provide a complete catalog of all acid deposition research and monitoring activities in the United States.

Research Progress and Outlook

MAJOR RESEARCH QUESTIONS

The National Program includes research, monitoring, and assessment activities to develop answers to major research questions. In addition to identifying and reducing scientific uncertainties, the National Program is organizing the scientific information to address policy questions. The Task Force is establishing a systematic framework for evaluating the costs and benefits of alternative policy options for controlling man-made sources of acidity or mitigating adverse effects.

Some of the overall research questions the National Program is specifically addressing are given here with some indication of the type of work underway to answer the questions.

1. **Does acid deposition significantly affect aquatic resources, crops, soils, forests, materials, or human health?** Except for poorly buffered lakes, evidence on other actual impacts is inconclusive. The National Program included intensive laboratory and field studies to develop a more definitive understanding of the potential for damage to sensitive resources.

2. **Where are the sensitive resources? How much damage has occurred, or how long will it take for damage to occur?** Concern about acid deposition centers around its potential and known effects. The National Program is developing inventories of the aquatic, terrestrial, and cultural resources at risk and is surveying the resources that have already been affected by acid deposition.

3. **What is the relative contribution of wet versus dry deposition acids?** Dry deposition may constitute up to 50 percent or more of acidifying substances, but reliable methods for its routine collection and measurement do not yet exist. Vigorous efforts are underway in the National Program to develop appropriate techniques for adequately monitoring dry deposition on a routine basis. In the meantime, interim methods for collecting dry deposition will be used to provide the best currently attainable data.

4. **What is the relative importance of local versus distant pollution sources in controlling the acidity of deposition at a given location?** Neither models nor measurements currently address this question with a reasonable degree of confidence. The National Program includes a broad atmospheric sciences effort with models, experiments, and measurements in the field and laboratory designed to improve our understanding of source/receptor relationships as rapidly as possible. Information on the characteristics and magnitude of local and regional sources of man-made emissions is being refinieded as well.

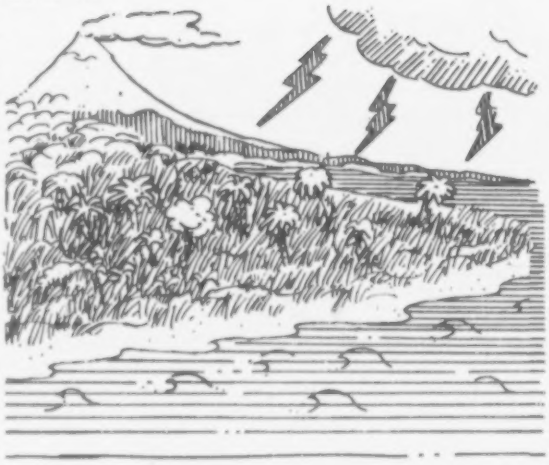
5. **What is the relative contribution of man-made versus natural sources of acid-forming material?** Significant natural sources exist, but their role is uncertain. The amount of natural acid precursors arising from sources such as oceans and marshes is being determined by the National Program to assess their contribution to acidity over North America. The magnitude, distribution, and character of man-made emissions are also being inventoried to more accurately identify their contribution to acid deposition.

6. **What is the temporal and spatial distribution of acid deposition?** Monitoring efforts before 1977 were fragmented and generally inadequate for determining trends. A National Trends Network with standardization and quality control has been established to answer these questions. Using a variety of information sources, research is also underway to improve monitoring techniques and to decipher any past trends.

7. **What changes in deposition would result from a given change in sulfur dioxide, nitrogen oxides, organics, and/or primary sulfate emissions?** Scientists cannot precisely calculate the amount of emission reductions that would be required to reduce acid deposition below a particular level within a given area. We need to know more about atmospheric processes to accurately quantify source/receptor relationships. The National Program's assessment activities integrate information on sources and atmospheric processes to address this question.

A. Natural Sources

BACKGROUND*



The sources of acid precipitation precursors are both natural and man-made. Natural sources will continue to contribute to precipitation chemistry despite any foreseeable abatement procedures that might control man-made sources. Emission control strategies and alternative energy options, therefore, require an accurate assessment of the relative importance of the two types of sources.

To make that assessment, the National Program's natural sources research focuses on answering two key questions:

- What natural emissions can influence precipitation chemistry?
- What are their strengths locally, regionally, nationally, and globally?

This research seeks answers to these questions in the context of current knowledge of natural emissions, which is summarized as follows.

The present understanding recognizes both biogenic and nonbiogenic natural sources. Some estimates place biogenic sources—vegetation and microbial activities in upland terrestrial sites, tidal/intertidal areas, and nutrient-rich oceanic regions—at 80 percent of the total natural source strength. The nonbiogenic sources include geothermal activities, natural combustion, lightning, and airborne soil and water aerosols. Sulfur- and nitrogen-containing gases are currently suspected to be the principal emissions from natural sources that influence precipitation chemistry, but natural alkaline materials such as ammonia and wind-generated aerosols and other acidifying components such as chlorine compounds could also play significant roles.

Various researchers have estimated numerous global budgets for sulfur compounds and have put natural emissions from 35 to 85 percent of the total global sulfur budget. On regional scales, the few existing estimates place natural sulfur emissions at a much smaller fraction, only a few percent in one study of the northeastern United States. However, these estimates are derived largely from a few measurement series. Furthermore, it is not clear whether differences among the data arise from natural variation within the environment or from problems with the instruments and techniques used in measuring natural sulfur emissions.

A recently completed assessment of nitrogen oxide emissions found that industrial and agricultural emissions together are approximately twice natural emissions on a global scale, while in North America fossil fuel combustion was found to produce 3 to 13 times more nitrogen oxide than natural sources. Systematic studies are required to accurately determine natural nitrogen oxide emissions with higher accuracy and geographical resolution than is currently available.

RESEARCH GOALS AND ACTIVITIES

To resolve these uncertainties, the National Program's research on natural sources has set the following specific objectives for FY 1982 to FY 1985:

1. Establish the reliabilities of the current state-of-the-art sulfur flux-measurement instruments and techniques.
2. Identify via field measurements, modeling, and laboratory studies the major potential sources of natural sulfur emissions that could contribute to precipitation chemistry.
3. Determine the strength of acid-forming natural sulfur emissions from land and ocean sources by using existing data and supplementary studies.
4. Develop accurate techniques for identifying and quantifying natural nitrogen emissions that could contribute to precipitation chemistry.
5. Estimate the strengths of acid-forming nitrogen emissions using survey measurements.
6. Provide an initial assessment, measurement technique development, and survey measurements of the natural alkaline emissions that can contribute to precipitation chemistry.

Reaching these goals will lead to a significant accomplishment by the National Program: an experimental data base that adequately characterizes the source strengths of the natural sulfur, nitrogen, chlorine, and alkaline emissions that influence precipitation chemistry on regional and national spatial scales.

* These background sections are based on scientific information available from a variety of sources, including research sponsored by Federal and State agencies, environmental, private sector, and international organizations, and other countries.

These findings, together with analogous ones from man-made sources research, will feed into the atmospheric transport/transformation, source/receptor models used to study atmospheric processes. The models will then be able to assess the impact of natural emissions relative to man-made emissions.

PROGRAM ACCOMPLISHMENTS—FY 1982

The research projects on natural sources began in FY 1982. Because of delays in budget allocations, funds became available midway through the fiscal year; hence, the results summarized here represent only the start of the program.

- **Instrument Development**. The initial research thrust in FY 1982 was to develop and test analytical techniques for sulfur- and nitrogen-containing species. One of the major problems in reliably measuring low sulfur concentrations, water vapor in the sample, has been satisfactorily solved by developing a membrane that selectively removes water without altering most other concentrations. A sensitive technique for measuring concentrations expected for natural nitrogen emissions has also been developed. Both of these instrumentation advances should greatly improve the quality of future field measurements required by the National Program's research in natural sources and atmospheric processes.
- **Modeling**. Studies are beginning to partition the likely sources of natural nitrogen compounds. Using measurements obtained in remote oceanic areas, photochemical models have shown that most of the nitrogen produced over the oceans arises from lightning, with about a 10 percent contribution from the stratosphere. In FY 1983, these studies will be extended to other areas to improve estimates of the contribution of lightning over continents.
- **Field Measurements**. Preliminary oceanic field measurements have found reduced sulfur compounds in biologically productive seawater in the Gulf of Mexico and the North Pacific Ocean. Crude estimates of release rates of these compounds to the atmosphere from the ocean suggest they could be a significant source of atmospheric sulfur. These data will guide the FY 1983 oceanic measurements described here.

FUTURE ACTIVITIES

A major element in the natural sources research program is determining the confidence levels of various techniques used to estimate sulfur gas emissions from natural sources. The information gained not only will permit a better assessment of the existing data base, but will help plan how to improve it: both efforts will take place during FY 1983 and FY 1984.

Also in FY 1983, the Natural Sources Task Group will comprehensively review the existing land-based sulfur emission data base, emphasizing the acid precipitation problem. This inventory, categorized by soil types, will yield a "first look" at the magnitude of the natural sulfur source budget, and it will be used to identify existing needs. Field measurements will occur in the following year as needed, drawing on the previous instrument development.

Parallel to these studies, a detailed field and laboratory investigation will examine the characteristics of specific major sulfur emitters. These latter data will provide by the middle of FY 1984 a much needed prognostic understanding of sulfur sources and their importance to the acid deposition problem.

Research cruises from FY 1983 to FY 1985 will further explore oceanic emissions of sulfur. Emphasis will be placed on quantifying the factors that influence the rate of fixation of inorganic sulfur by marine organisms, their release of reduced sulfur compounds and the vertical distribution of these compounds within the upper oceanic layer, and the transport of reduced sulfur compounds across the sea-atmosphere interface. Additionally, the correlation between the areal distribution of reduced sulfur compounds in productive seawaters with biological parameters that are amenable to remote sensing will be determined.

Laboratory kinetic studies will investigate the paths by which natural emissions are converted to acids. These rates and mechanisms are necessary in atmospheric models that will assess the acidification of precipitation by natural emissions. Initial estimates of sulfur compounds will begin in FY 1984 and continue through FY 1985.

In FY 1983 distributions and deposition fluxes of nitrate, sulfate, and ammonium will be modeled over remote oceanic stations, where the understanding of natural processes can best be tested. It is anticipated that these studies will point out previously unsuspected sources, if they exist. An initial assessment of the global budget of atmospheric nitrate precursors will be made in FY 1985.

The study of natural alkaline materials will be completed in FY 1983. This research will gather existing data on airborne alkaline materials; assess amounts and identities by region, seasonal variation, influence of meteorological conditions, and solubilities; and thereby estimate their potential neutralizing effect on precipitation.

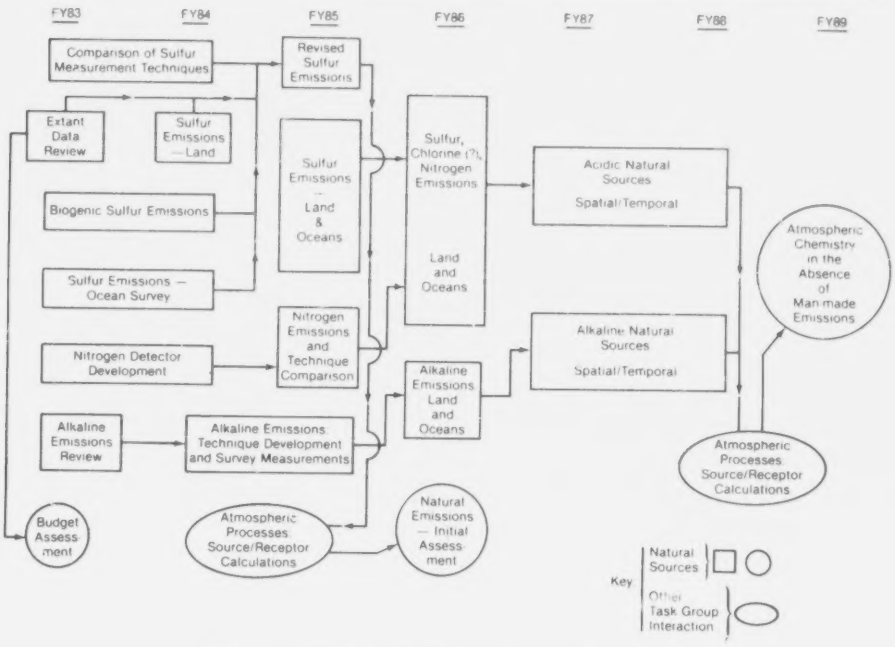
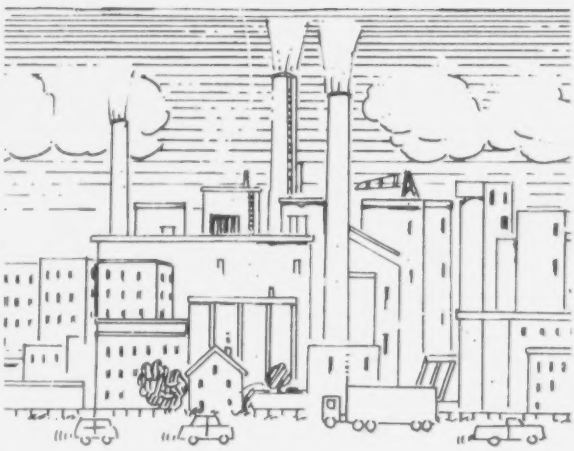


Figure A.1.—Proposed program implementation strategy, Natural Sources.

B. *Man-Made Sources*

BACKGROUND



Research on acid deposition has identified several substances believed to be precursors to the formation of acid compounds in the atmosphere. As more knowledge of the acid formation process is gained, the list may change.

A current list of pollutants likely to be important includes sulfur dioxide, oxides of nitrogen, volatile organic compounds, primary sulfates, ammonia, hydrochloric and hydrofluoric acids, and alkaline dusts. Of these, sulfur dioxide and oxides of nitrogen have long been recognized as important; studies of the potential influences of other pollutants, including trace metals, are continuing.

Experimental work is developing emissions factors, its purpose is to increase the accuracy of existing factors and to develop new factors for pollutants recently identified as potential precursors of acid deposition. Typically these factors are expressed as pounds of pollutant emitted for a unit produced or unit of fuel consumed. The quality of any emissions inventory depends on the accuracy of these factors. Work to develop new emissions factors is focusing on nitrogen oxides, primary sulfates, hydrocarbons, hydrochloric acid, and trace metals such as vanadium and manganese. Emissions factors for sulfur oxides are being refined.

Total emissions of sulfur dioxide (SO_2) and oxides of nitrogen (NO_x) for the United States and Canada in 1980 were calculated as part of the work done for the U.S./Canadian Memorandum of Intent. These calculations are (in million tonnes per year):

	United States	(%)	Canada	(%)	Total
SO_2	24.1	(55.5)	4.8	(72.7)	28.9
NO_x	19.3	(44.5)	1.8	(27.3)	21.1
		(100.0)		(100.0)	

Canadian emissions of nitrogen oxides make up a smaller percentage of that country's total emissions than do domestic NO_x emissions in the U.S. total. A large portion of the Canadian sulfur dioxide emissions is generated by primary metal smelting, which contributes relatively few nitrogen oxides.

Over the past 60 years U.S. sulfur dioxide emissions have fluctuated between about 16 and 28 million tonnes per year, primarily following the rate of coal use in the country. The low point in total emissions was during the 1930's, the high point during World War II—closely followed by the early 1970's, before the 1970 Clean Air Act had an impact. The bulk of these emissions in the United States occurs east of the Mississippi River.

In contrast, nitrogen oxide emissions have increased more evenly over the years in response to increasing fuel use. The shift in coal use from domestic and industrial grade combustion to utility pulverized coal combustion has also increased average emissions because of higher combustion temperatures.

Future projections of sulfur dioxide and nitrogen oxide emissions depend on assumptions about future economic activity and regulatory changes. With no changes in regulation, the U.S./Canadian Work Group projects a 7.8 percent increase in sulfur dioxide emissions (a 5 percent drop in Canada offsetting a 10 percent increase in the United States). The Work Group expects nitrogen oxide emissions to increase a total of about 25 percent. Different retirement rates of older industrial facilities in the United States and replacement by new facilities subject to the existing emissions limitations for new sources could make these preliminary estimates highly uncertain.

RESEARCH GOALS AND ACTIVITIES

The fundamental purpose of the National Program's man-made sources research is to provide the other Task Groups with information on man-made pollutants needed for the overall assessment program. This research involves developing estimates of past, present, and future emissions of pollutants from man-made sources and then devising quantitative methods to estimate the effects and costs of possible government regulation of these emissions. The overall purpose of this research effort can be summarized in the following objectives:

1. Provide, with adequate geographic, temporal, and economic sector resolution, an accurate and complete inventory of emissions from man-made sources believed to be important in acid deposition processes.

2. Provide models that (a) predict how these emissions may be altered by such factors as economic growth, fuel supply, emissions regulations, and control techniques, and (b) calculate the cost of alternative control strategies.

A series of projects is being conducted to attain these research objectives. In 1985 an interim version of the emissions data required to achieve these objectives will be completed, and in 1986, an interim group of models. The Man-made Sources Task Group plans to submit final versions of the emissions data base and the required models in 1988.

PROGRAM ACCOMPLISHMENTS—FY 1982

One major accomplishment in FY 1982 was to adopt an organizational structure to compile and analyze research results. In addition, continuing technical projects have provided valuable information on meeting National Program objectives and on conducting future projects. Major accomplishments are summarized.

- **Preliminary General Specification for the Man-made Emissions Inventory Data Base**. Current work is delineating the pollutants to be included in the inventory's data base, specifying the geographic resolution, the time frame (annual, weekly, daily), and the sources of pollutants for which emissions data will be provided. This will set a uniform standard for emissions data to be used by the National Program in examining atmospheric processes related to acid deposition. All man-made sources research projects will be evaluated in terms of their ability to supply necessary data, and, if needed, will be redirected to satisfy National Program needs.

- **Preliminary General Specification for the Man-made Sources Models**. These models will provide a tool to help analysts develop and assess various regulatory strategies. The National Program requires specific data to be generated by man-made sources models, given general descriptions of various projected emissions levels. The models will help determine potential decreases in emissions from a set of policy options and their costs. The Man-made Sources Task Group will review the specifications for the models, then transmit them to the scientific community for review and comment. The final specification will be used to evaluate existing and planned projects and make appropriate adjustments.

- **Initial Plan for Acid Deposition Inventory**. This document proposes a plan for creating a data base for the emissions inventory, which will provide a single source of emissions data within the National Program. The inventory is designed to meet the information needs of National Program researchers and other interested persons in the scientific community who are explaining and modeling atmospheric processes related to acid deposition and developing and evaluating pollution reduction techniques.

- **Acid Precipitation Precursor Study**. The completed Acid Precipitation Precursor Study presented findings on the general relationship between fossil fuel use and the location and degree of acid precipitation. This study points out the potential importance of petroleum combustion in addition to coal combustion in forming acid deposition. The National Program will use this document to help plan its emissions inventory activities, including the potential expansion of the emissions examined and the definition of required resolution. As a result of this study, the National Program will also examine emissions from transportation activities more closely.

- **Studies Characterizing the Sources of Acid Precipitation**. Studies begun in the latter part of FY 1982 and continuing through FY 1984 include (1) investigation of boron as a tracer of aerosol from the combustion of fossil fuels; (2) sulfur isotope analysis; (3) receptor modeling, precision, accuracy, and data requirements; and (4) source characterization for receptor model source apportionment of acid rain precursor species. Because these studies have begun recently, they have not yet produced results for publication. Eventually, they will lead to improved characterization of sources of acid deposition precursors.

All these accomplishments reflect the emphasis in the latter half of 1982 to better define the products to be provided by the Man-made Sources Task Group to other research groups, and to formulate effective and economic plans for meeting these needs. This effort will provide more accurate and timely results with a minimum of expenditure.

- **Advanced Utility Simulation Model (AUSM)**. When completed, this model will allow the National Program to evaluate the costs and changes in emissions that would result from a wide range of control strategies for utilities. In FY 1982 a prototype, single-State version of the AUSM was developed and successfully operated for several States. A data base was developed that will allow operating the model for each State. The AUSM can be used with other models to compare costs and emission changes for strategies involving different combinations of sectors, pollutants, and regions. This modeling effort used in conjunction with the other emissions models will show how emission patterns might be altered by a number of control scenarios and the cost of each scenario. These emission pattern data coupled with transformation/transport models and acid deposition/damage models will help determine the cost/benefit relationship for the control scenarios that might be considered.

- **Industrial Combustion Emissions Model**. Work was begun on this model, which, when completed, will be able to evaluate the costs and emissions reduction potential of alternative regulatory strategies for industrial combustion sources. The model will focus initially on the industrial boiler sector.

FUTURE ACTIVITIES

The National Program's Man-made Sources Task Group's major goal in FY 1983-84 is to review and modify specifications for the Man-made Emissions Data Base and Model Set. After completing this in early FY 1983, the group will review its own projects against the specifications to ensure they satisfy National Program needs.

Besides working to establish the Man-made Emissions Data Base, the group will continue work on the utility and industrial emissions models during FY 1983-84. Under the National Program, man-made sources research will develop a comprehensive emissions inventory for a 1980 base year. This inventory will separate information on

sulfur dioxide, nitrogen oxides, sulfates, and volatile organic compounds on a seasonal basis for the United States and southeastern Canada, covering both point and area sources. Research will develop emissions factors for ammonia, chlorides, fluorides, and alkaline dusts.

Planning will continue for compiling an improved emissions inventory for a 1984 base year. Finally, the group will develop an historical inventory of sulfur dioxide and nitrogen oxides.

Advanced Utility Simulation Model. In FY 1983-84 a fully documented State-level version of the AUSM and associated data bases will be transferred to government computer systems and tested. In addition, work will begin on developing the remaining components required to operate the model for either national or regional analyses.

Industrial Combustion Emissions Model. The effort to develop the Industrial Combustion Emissions Model will intensify, with delivery scheduled for early in FY 1984. FY 1983 activities will focus on modifying the fuel choice decision module. Process heaters may be added to the model's capabilities. The Technical Advisory Committee will continue to guide the project's activities and evaluate its products.

Industrial Process Emissions Model. The Environmental Protection Agency will begin to develop an Industrial Processes Emissions Model in FY 1983, accelerating these activities through FY 1984. Initial emphasis will be on identifying the most significant pollutants and the industries emitting them, along with determining the best mechanism to model emissions from industrial processes. The model will be able to evaluate the cost and effectiveness of alternative control strategies imposed on industrial processes. This model will be compatible with the Industrial Combustion Emissions Model, thereby aiding analyses of the entire industrial sector.

The refinement process for these models will be undertaken in FY 1985 and continue through FY 1988.

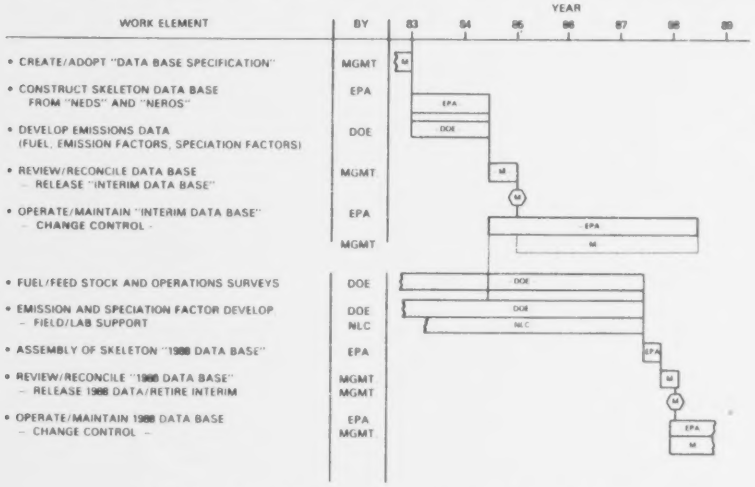


Figure B-1.—Proposed schedule—emissions data base, Man-made Sources.

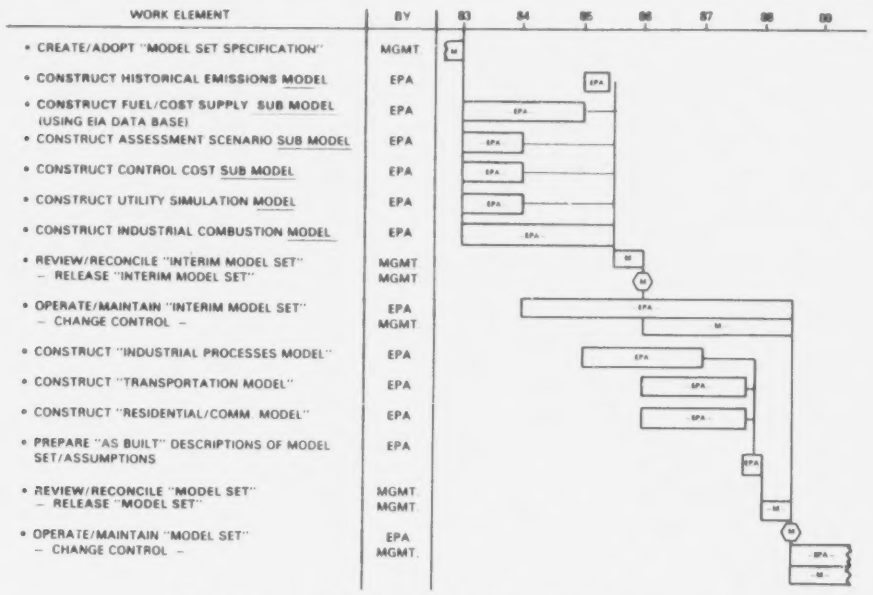
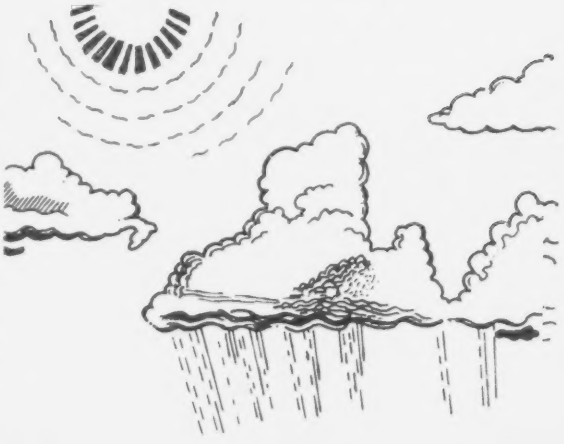


Figure B.2. Proposed schedule - model set data base, Man-made Sources.

C. *Atmospheric Processes*

BACKGROUND



One of the basic observations about acid rain is the approximate coincidence of the region of highest precipitation acidity with the areas of greatest sulfur dioxide and nitrogen oxides emissions. Further, we know that these two pollutants are the primary precursors to acid materials, and are transformed in the atmosphere, at least partially, to sulfuric and nitric acid. To date, however, no one has been able to reliably assess the contribution of a specific source or set of areal sources to the acidity of deposition in a given distant region. To establish this source/receptor relationship, which is needed to evaluate the effectiveness of different proposed control strategies, the relationship between emissions and deposition must be reliably quantified.

Research on atmospheric processes is subdivided into three major categories:

Transport. The speed and direction of the winds at the level of ejection are the major influences on the movement of the pollutants. How far they are moved depends upon whether the given pollutants are gases or particles, at what level they enter the atmosphere, and how they are transported vertically during their lifetime. For example, gases emitted at or transported to levels well above the earth's surface will travel farther than large particles ejected at the surface. Such factors are important in understanding local versus long-range transport.

Transformation. While the material is being moved by the air currents, chemical changes can take place. For example, smog may form in the presence of sunlight, or sulfur dioxide may be incorporated in a cloud droplet, forming sulfuric acid. These complex interactions, especially those in clouds, are not yet well understood and probably hold an important key to explaining acid deposition patterns.

Deposition. After the pollutant material is transported and possibly chemically transformed, it is finally deposited either by contacting a surface directly (dry deposition) or by being incorporated into precipitation and brought down to the earth's surface in rain or snow (wet deposition). The relative importance of wet and dry deposition is one of the major questions still unanswered.

Because transport, transformation, and deposition form such a complex system, scientists have tried to sort out their relative importance by using mathematical models. At present, the models cannot account for every step but have been designed to cover the gross features of the atmospheric processes. The major emphasis to date has been on transport studies, with very rough approximations of the transformation and deposition processes.

Plans are being made to incorporate more realistic conditions into future, more sophisticated models. A major goal of the National Program's atmospheric processes research is to develop reliable models that can be used in a predictive and interpretive mode to explain source/receptor relationships.

Because the development of reliable models is complex, alternative approaches to determining source/receptor relationships are being developed. One way would be to conduct a series of field experiments in which inert tracers or isotopically tagged man-made pollutants released into the atmosphere are measured and then tracked while they are transported and diluted, transformed physically and chemically, and eventually deposited on the earth's surface. Such studies provide empirical information on the processing of these pollutants between sources and receptors.

RESEARCH GOALS AND ACTIVITIES

The National Program's research on atmospheric processes is designed to provide an understanding of the atmospheric link between the emission of pollutants and acid deposition. Understanding this link requires research that addresses the following key questions:

1. What are the important chemical species and processes governing the formation of acidic substances in the atmosphere and how do they operate?
2. What are the most important meteorological processes governing the transport of acidic substances and their precursors?
3. How do the processes of wet and dry removal operate and how does their relative importance change as distance from the source increases?

4. How can the processes of transport, transformation, and removal be modeled so that we can understand current and predict future deposition patterns?

The answers to question 1 will tell us which atmospheric pollutants will have to be controlled to reduce acid deposition. The answers to questions 2 and 3 will indicate which source regions are most likely to affect a given receptor region. However, the models developed in response to question 4 will be the primary policy development tools. These models will provide quantitative estimates of the effects that changes in existing pollution sources will have on existing patterns of acid deposition. In other words, the models will tell us what the pollution sources have affected, what changes (if any) must be made, and in what quantities.

To answer these questions, the atmospheric processes research focuses on the following specific objectives:

- Improve understanding of the vertical and horizontal transport of acidic substances and their precursors.
- Improve understanding of the major chemical transformations that produce acidic substances in the atmosphere.
- Improve understanding of precipitation scavenging processes in the atmosphere.
- Improve treatment of dry deposition in numerical models of source/receptor relationships.
- Improve and develop predictive and interpretive models that explain source/receptor relationships in the atmosphere.
- Perform field studies that are necessary to provide a data base for developing and testing source/receptor models. Such studies will also provide an independent way of evaluating the source/receptor relationship on an empirical basis.

PROGRAM ACCOMPLISHMENTS—FY 1982

Transport

- **Air trajectories were calculated from the receptor of acid deposition backwards in time for numerous sites in the United States.** This technique allows a chemical climatology to be established for various areas in the United States. Initial results have been published, and the work continues on a routine basis. The importance of connecting the deposition chemistry with the transport meteorology is one of the necessary activities in understanding patterns of acid deposition and their impact on sensitive ecological areas.

- **Data from the sites in the Global Background Research Program are being used to evaluate the long-range transport on a hemispheric scale.** Results show that acidity of sites in remote areas is higher than would be expected if carbon dioxide were the only factor influencing pH at "clean air" sites. Weak and strong acids found in the samples show that the total acidity may be related to a mixture of natural and man-made sources. These results will be used in subsequent model calculations.

- **Initial observations confirm the importance of clouds in the transformation process of precipitation chemistry.** Research results support the hypothesis that fair weather cumulus clouds play a major role in transforming sulfur dioxide to sulfate during the late spring, summer, and early fall. Aircraft measurements of air pollutants above and below fair weather cumulus clouds indicated that these clouds transported pollution out of the sub-cloud boundary layer and into the air above. Moreover, the air pollutants processed through the clouds underwent significant chemical transformation.

- **A large cooperative program was started to study the eastern North American chemical budget.** Since August 1982 numerous private and government organizations including NOAA, NASA, General Motors, and others have cooperated in a study that consists of intensive measurements of aerosols both on the ground and by aircraft, precipitation collection and analysis, and meteorological flow studies. The results of this project will help define one portion of the sulfur and nitrogen budget of eastern North America, that is, what percentage of these substances are transported off the East Coast.

- **Planning and initial testing of field equipment began for Cross Appalachian Tracer Experiment (CAPTEX).** This long-range tracer experiment, which will begin in 1983, consists of releasing an inert tracer under specific meteorological conditions and measuring the transport of the tracer over hundreds of miles. The results of such experiments will be used to help verify meteorological models.

Transformation

- **Preliminary results were obtained from a study of the influence of man-made materials on the cloud-forming process.** Measurements made of clouds and aerosols in polluted and nonpolluted air in the Northeast identified man-made aerosols hundreds of kilometers downwind from the source. This study is a part of the effort to understand the chemistry and scavenging processes in the atmosphere, and will provide necessary input for model development.

- **Preliminary results were obtained from a laboratory study investigating the kinetics and photochemistry of nitrate radicals.** A laboratory experimental apparatus has been designed and assembled to study the chemistry of the nitrate radical. This radical is an important member of the nitrogen oxide family and is involved in both gas-phase and heterogeneous reactions leading to nitric acid formation. This experiment will provide critical data needed to model the chemistry of nitrogen oxides, particularly in urban environments, and will provide direct input to the more sophisticated models planned for FY 1985. In a separate experiment, kinetic data have been obtained on the self reaction of hydroperoxyl radicals. This reaction is very important because it produces hydrogen peroxide, thought to be the major oxidizer of sulfur dioxide to sulfuric acid.

- **The atmospheric transformation of nitric oxide, nitrogen dioxide, and nitric acid in the clean marine atmosphere is being investigated to increase our understanding of the chemistry that produces nitric acid.** The atmospheric photochemistry of the oxides of

nitrogen has been modeled for clean marine air. The oxidation of nitrogen dioxide to nitric acid by the hydroxyl radical in the gas phase is probably a major mechanism in converting nonacidic oxides of nitrogen to nitric acid in rural environments. The research results will be useful in evaluating the oxidation rate of nitrogen oxides in the relatively unpolluted regions of the atmosphere.

Deposition

• **Reanalysis of the past precipitation chemistry data suggests the recent trend toward increased precipitation acidity in the midwestern United States is more a consequence of less alkaline dust in the precipitation than greater concentrations of acidifying agents.** The complexity of interpreting past precipitation chemistry data to establish trends of acid deposition has been a topic of controversy mainly because of the unknown quality of the data. Recent information from federally funded research emphasized the necessity for total chemistry interpretation, not just focusing on the isolated acidic components. A rational interpretation of the chemistry network data of the mid 1950's suggested that drought-induced increases in alkaline crustal dust decreased precipitation acidity.

• **The first comprehensive descriptive maps of the distribution of major chemical species over North America have been produced.** The maps, prepared from the MAP3S, NADP, and CANSAP data bases, display seasonal concentrations and depositions of calcium, ammonium, nitrate, sulfate, and hydrogen ion. In general, the maps show relatively high concentrations of acidic species over the upper northeastern States and lower Canadian Provinces, but the greatest acid deposition is wholly confined to the United States over eastern Ohio, western Pennsylvania, and northern West Virginia.

• **Research on deposition of trace elements in forests indicates that atmospheric deposition dominates the landscape cycle of lead and has a measurable influence on the cycles of cadmium and zinc, but a minimal influence on the manganese cycle in a deciduous forest in the eastern United States.** Rain event deposition rates are orders of magnitude greater than the intervening dry deposition rates; however, dry deposition supplies 20 to 90 percent of the total annual input to the forest. This research directly links atmospheric deposition, both wet and dry, and the effects research on terrestrial ecosystems. The National Program recognizes that the study and sampling of atmospheric deposition of metals should not be overlooked, and that dry deposition must be included in future network sampling strategies.

Models

• **A new wet scavenging model has been used to analyze precipitation data from the recently completed Oxidation and Scavenging Characteristics of April Rains study (OSCAR).** The model includes an improved treatment of the physical process of scavenging and an improved, nonlinear chemistry module that accounts for aqueous phase reactions of sulfur and nitrogen compounds. In an application to one of the OSCAR storms, the model was able to explain the chemical composition of a sequence of precipitation samples. The model strongly indicates that scavenging rates for that particular storm were governed by the concentrations of oxidants such as hydrogen peroxide and ozone and not by the ambient concentrations of sulfur and nitrogen oxides. Understanding the physical and chemical processes governing wet deposition is important in developing improved source/receptor models and determining the emissions that may have to be controlled in any action to mitigate acid deposition. The results from the OSCAR model will be integrated into the overall atmospheric processes modeling effort.

• **The Advanced Statistical Trajectory Regional Air Pollution Model (ASTRAP) was used along with seven other linear source/receptor models to analyze wet deposition measurements in the eastern United States and Canada for the year 1978.** A procedure was developed for using deposition monitoring data to evaluate source/receptor models; however, the actual evaluation showed that there was an insufficient data base for determining whether or not existing source/receptor models are capable of explaining current deposition patterns. Since 1978, the monitoring effort has expanded, especially in the Northeast. From 1980 to the present, an excellent data base exists that the models can be tested against.

FUTURE ACTIVITIES

Research under the National Program will continue to improve our understanding of atmospheric processes; particular emphasis will be on improving the ability to determine source/receptor relationships so that the consequences of changes in emission can be reliably predicted.

Highlights of the expected accomplishments for FY 1983 to 1985 include:

A comprehensive plan for developing an advanced nonlinear source/receptor model will be produced and implemented. Further advancement in modeling through such nonlinear models will be critical in planning regulatory options.

Work will begin on a detailed experimental and management plan for conducting a major field experiment to provide data for developing and evaluating source/receptor models. The planning for field activities will begin in FY 1983 with possible implementation in FY 1985.

The Cross Appalachian Tracer Experiment will be conducted, providing the first sizeable data set for evaluating long-range trajectory models. To do this, inert tracers will be released in the Midwest and in Canada, and their concentrations measured over 800 miles away on the East Coast. How well the models' predictions compare to the actual measurements will help put limits on the usefulness of the models. The first major CAPTEX will be in September to November, 1983. Based on the results of that experiment, CAPTEX II will be planned for FY 1985.

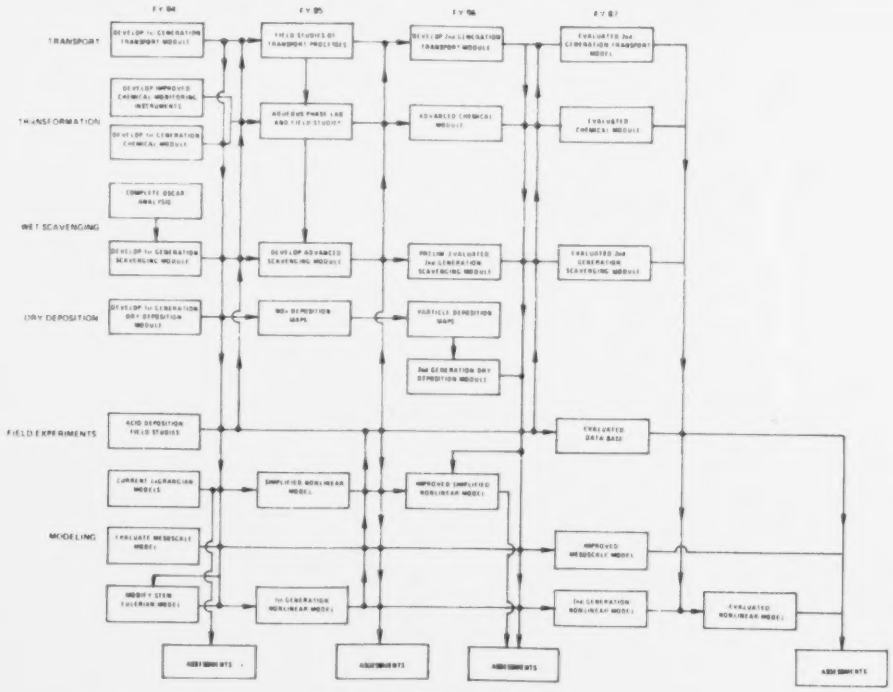
In FY 1983 research to investigate the source/receptor relationships of long-term precipitation chemistry data bases at Hubbard Brook and from the MAP3S network will be completed. These studies will demonstrate the feasibility and limitations of the trajectory analysis ap-

proach defining source/receptor relationships, and will be used to evaluate the more complex models.

The North American sulfur and nitrogen study will, by the end of FY 1983, produce preliminary results on amounts of materials transported out to sea. These initial estimates of the sulfur and nitrogen transport will be used in the National Program's work on assessments and policy analysis.

The atmospheric trajectory models will be used in various parts of the world for precipitation chemistry evaluation. By the end of FY 1984, the Southern Hemisphere data will be added so that trajectory calculations can be made there also. This will allow scientists to evaluate the global transport of acid-forming materials.

The key reactions that control the atmospheric production and loss of hydrogen oxides and ozone will be identified and measured. Thus, the gas phase oxidation of sulfur dioxide and oxides of nitrogen can be quantitatively estimated. A review of the current acid deposition models and the draft plan for developing a Eulerian acid deposition model will be completed to determine how information on the critical reactions can be incorporated in these models. This study will be finished in FY 1983. The recommendations will be used to direct and focus this modeling effort in FY 1984 and 1985.

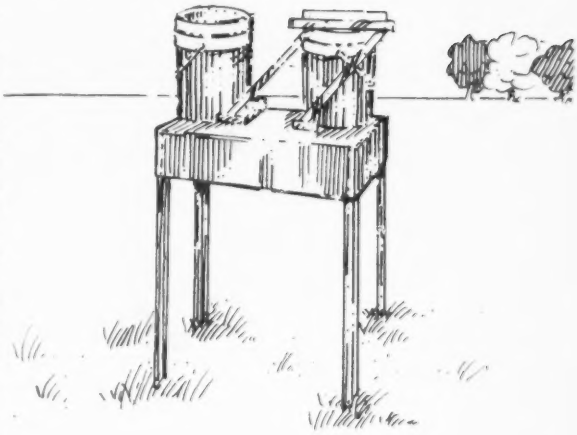


TASK GROUP C: PROGRAM LINKAGE

Figure C.1. Proposed program implementation strategy, Deposition Monitoring.

D. *Deposition Monitoring*

BACKGROUND



Precipitation chemistry in the United States has been measured on an erratic basis since the early 1920's. These early data were gathered mostly for agricultural research. Since the early 1970's careful attention has turned to the design and operation of collectors and networks, and to procedures that will improve the data quality.

Monitoring of dry deposition is in its infancy. Although collectors trap wet and dry materials separately, little credence is given to the dry portion because gases are not collected and the collecting surface (a plastic bucket) does not replicate the natural surfaces of vegetation, soils, and waters. Thus, present estimates of dry deposition are based on limited research results under specialized, short-term conditions.

The two major networks for wet deposition in North America are the National Atmospheric Deposition Program (NADP) in the United States and the Canadian Network for Sampling Precipitation (CANSAP) in Canada:

	Start date	No. of sites	Frequency of sampling	Type of collector
NADP	1978	108	weekly (wet) and bi-monthly (dry)	wet & dry
CANSAP	1977	59	daily, monthly composite	wet only

The distribution of annual average pH of wet deposition on North America in 1982 is shown in Figure D-1. Isopleths have not been drawn for the western part of the continent because abrupt changes in topography in mountainous areas require the development of modified procedures for their construction. The regions of lowest pH (4.2) in eastern North America roughly coincide with the regions of highest man-made emissions of acid precursors (primarily sulfur dioxide) in the midwestern and northeastern United States and near the Sudbury smelters in Canada. The continuing pattern of lowest pH values in the northeastern United States and Canada suggests that the impacts of acid-forming emissions are felt most strongly within the regions where those emissions are produced. This does not suggest that longer-range transport is unimportant, however.

Variations of 30 to 50 percent in pH values sometimes occur among sites within a given State. This may indicate effects of local sources, insufficient quality control, or both. These variations underscore the importance of long-term data sets in describing geographical trends. Also, most NADP/National Trends Network sites are in rural precipitation in the United States. Because of the presence of many local emission sources, many studies have found precipitation in urban areas to be more concentrated in acid-forming constituents, as a general rule, than precipitation collected in rural areas.

Network data also show that concentrations of many pollutants tend to be high when precipitation volume is low. Thus, fogs and mists are more acidic than heavy rains. This may be important in assessing damage to pH sensititve receptors.

Values of pH substantially less that 5.6 have been reported in some remote areas of the world where annual average pH values have been found to be as low as pH 4.9. Such measurements have demonstrated that a pH of 5.6, often considered "normal" for wet deposition in the absense of man-made, acid-forming gases, cannot be considered a benchmark for identifying regions affected by man-made emissions.

In North America, historical trends in precipitation chemistry have been difficult to quantify because of differences in past collection methods and sampling sites, the brevity of the records available, and the relatively modest changes in precursor emissions in recent years. The limited data available from past studies suggest that calcium concentrations may have decreased, nitrate concentrations probably increased, and sulfate concentrations apparently changed little in the past 25 years. Data from the mid-1950's show substantially higher alkalinity than contemporary data, perhaps because of a drought in the Midwest at the time. Recent studies reported in the literature also indicate that pre-1960 alkalinity values may have been overestimated by the analytical methods in common use that the time.

There are no widely accepted methods for monitoring dry deposition on a routine nationwide basis. The limited data available suggest that wet and dry deposition account for roughly equal amounts of acid deposits nationwide, with more dry deposition in arid climates, and less in humid areas. Near emission sources, dry deposition is believed to be a more important contributor of acid-forming substances than wet deposition because of rapid

settling of larger particles. Much additional research is needed on dry deposition measuring methods; a great deal of this work is now being done by agencies in the Deposition Monitoring Task Group.

The monitoring networks begun since 1977 have developed a fairly complete initial picture of wet deposition chemistry on a regional and national scale. But, sizeable areas of the western United States are not yet adequately covered, and the length of record is not yet sufficient to determine long-term trends. As additional sites are added to the National Trends Network and the record of each site becomes longer, a more complete, consistent, and long-term data base will become available. At present, subregional scale wet deposition patterns are less well delineated, especially near sources and in urban areas. Local variations in acidity are generally not well-defined. Variations in the chemical composition of wet deposition around local sources require intense study and monitoring to delineate. Studies of this sort are being conducted in a few urban centers, with more planned in future years.

Recent temporal trends in the chemistry of wet and dry deposition are difficult to quantify reliably, with the possible exception of the nitrate content of wet deposition, which shows increases roughly paralleling the increase in man-made emissions of nitrogen oxides. The difficulty in discerning recent trends is due to the shortness of the data record at most stations.

RESEARCH GOALS AND ACTIVITIES

Planning and coordinating atmospheric deposition monitoring under the National Program is the responsibility of the Task Group on Deposition Monitoring. The goals of the National Program's deposition monitoring activities are:

1. Determine the spatial and temporal variations in the composition of atmospheric deposition within the United States through a National Trends Network.
2. Develop methods for the reliable measurement of dry deposition.
3. Continue and increase research on methods for deposition monitoring.
4. Support a Global Trends Network (GTN) through operating monitoring sites at remote locations throughout the world.

Each of these objectives was emphasized in FY 1982.

PROGRAM ACCOMPLISHMENTS—FY 1982

• **National Trends Network.** The design of the NTN was completed. Recommendations for locations of 150 sites in 48 States (Figure D-2), were based on "Ecoregions of the United States" established by the U.S. Forest Service with consideration given to concentration gradients and sensitive areas. Approximately 90 existing NADP monitoring sites will be incorporated into the NTN. A site review team began visiting each existing and proposed NADP/NTN site and will issue approval or recommend changes for all sites by the end of FY 1983. The NADP/NTN is supported by a combination of Federal, State, and private funds and will continue to be coordinated as part of the National Acid Precipitation Assessment Program. Data from NADP/NTN will be used by other Task Groups as background for their studies and, in particular, by the Assessments and Policy Analysis Task Group for their assessments.

Figure D-1. Precipitation-weighted annual average pH of wet deposition, 1981. (Based on data from the monitoring network of the National Atmospheric Deposition Program and from the Canadian Network for Sampling Precipitation.)

Sufficient data are now available from the NADP network and the Canadian Network for Sampling Precipitation to permit an analysis of the spatial and short-term temporal trends in wet chemical deposition in North America. A 1982 NADP report evaluating pH data from these two networks through 1980 shows little apparent change in the distribution of acidity in the United States over the last 7 or 8 years, although there are few data from the early period. Data from NADP confirm that assessing future trends requires long-term monitoring to overcome year-to-year variations.

• **Global Trends Network.** Within the GTN (Fig. D-3), precipitation samples are collected in remote areas of the world and returned to the United States for analysis. Techniques developed in the GTN will be used in the operations of the NTN, while research-related data from the GTN will provide information on background levels of precipitation acidity for input into model calculations and for evaluating the overall acid rain problem. The GTN was established in part to measure precipitation chemistry in areas far from man-made sources; such measurements have shown an average rain acidity pH of about 5.0 at remote sites in Hawaii and Amsterdam Island in the Indian Ocean. It is anticipated that continued sampling from this program will show trends of global acidity and help describe the chemical climatology of different regions of the earth.

• **Quality Assurance.** The quality assurance program audits laboratory performance by regular submission of blind samples. Field measurement quality is

checked by semiannual submission of performance samples for analysis by over 100 site operators. Additionally, an experiment conducted in cooperation with North Carolina State University is measuring the effects of rain sampling frequency and within site variability on the precision of sampling atmospheric deposition data. The results of this experiment are expected to contribute to the assessment of the validity of using NTN data in describing regional patterns of acid precipitation.

At six border locations (three in Canada and three in the United States), U.S. and Canadian scientists collect precipitation samples according to both countries' protocols. The data from each collector at each site are compared to detect differences arising from collection techniques. This information will be extremely important when trying to evaluate possible cross border differences in chemical composition and validity of combining NADP/NTN and CANSAP data for trend analysis or other purposes.

• **Dry Deposition Methods.** In June 1982, an intercomparison field study of dry deposition monitoring and measurement methods was conducted for the National Program by the Illinois State Water Survey. Thirteen laboratories from the United States and Canada compared four micrometeorological methods, three types of surrogate surfaces, concentration monitoring, leaf washing, and material effects measurement. The analyzed data will be reported at a symposium in the spring of 1983.

Current plans call for measuring ambient air concentrations of relevant substances at monitoring stations nationwide. During FY 1982, a pilot effort was begun to compute dry deposition fluxes from air concentration and deposition velocities. Instrument shelters were installed at three locations representative of the surface types of the northeastern United States, especially forests and croplands. These sites will provide opportunities for applying detailed research techniques for direct flux measurement and for evaluating the deposition velocity calibration to be used at "concentration monitoring" sites for a future dry deposition network.

• **Data Base Management and Data Analysis.** The acid rain data base has grown during the year to over 100,000 observations in the United States and Canada. The networks currently entering data into the data base include NADP/NTN, CANSAP, MAP3S, and EPA's Region IV Network. EPA has developed an Interagency Agreement with Pacific Northwest Laboratories to design a more versatile Acid Deposition Data System to meet varying information demands.

Data analysis in FY 1982 included a prototype summary report on national acid deposition data trends and

Figure D.3 — Location of Global Trends Network (GTN) stations.

Figure D.2 — Locations of proposed National Trends Network (NTN) stations.

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an investigation of new methods for spatial analysis. The report examined new ways of presenting and simplifying the large amounts of data developed in a monitoring network, a technique for analyzing the confidence limits of the spatial distribution of given concentrations, and objective improvements in network design.

- **TVA Regional Trend Monitoring Network**. During FY 1982 TVA continued to operate three regional air quality trend monitoring stations. In addition to collecting biweekly wet and eight weekly dry deposition samples, these stations also monitor background concentrations of suspended particulates, suspended sulfate, sulfur dioxide, nitrogen dioxide, ozone, and gaseous flouride. Rigorous quality assurance/quality control procedures were implemented at the TVA central analytical laboratory for deposition analysis. A technical report "Atmospheric Deposition in the Tennessee Valley: 1979-1980" was published and presented at the 75th Annual Meeting of the Air Pollution Control Association. Plans call for upgrading one or more of these sites for inclusion in the NTN.
- **New Research Sites Established**. Three research sites will conduct special measurements and test newly developed equipment and methods for monitoring wet and dry deposition. These sites will also serve as the standards for their regions. They are located at Pennsylvania State University, the Walker Branch Watershed in Oak Ridge, Tenn., and Niwot Ridge in a biopreserve in the Rocky Mountains in Colorado.

FUTURE ACTIVITIES

In FY 1983 the Task Group will concentrate on implementing most of the sites of the NADP/NTN, developing methods for monitoring dry deposition, and further improving methods for monitoring wet deposition. Quality assurance programs will be used to oversee the data collection and analysis to assure data comparability among the several operational and research networks.

National Trends Network. Coordination and operation of the NTN will continue , including establishing 20 to 40 new sites in cooperation with several Federal, State, and private sector agencies. Site visitation and evaluation of all existing and proposed NTN sites will be completed and the agencies named to operate those sites started in FY 1983 and FY 1984. The Task Group will produce a special study of the Global Trends Network data using both recent and historical data.

Development of Dry Deposition Monitoring Methods. Intercomparison field studies will be run, one each at three core research sites in Oak Ridge, Tenn., Argonne, Ill., and State College, Pa. *Development of Flux Monitoring Methods:* The performance of a variety of micrometeorological methods will be tested and compared with alternative techniques such as surface accumulation. *Development of Concentration Monitoring Methods:* The temporal distribution functions for concentration and velocity of deposition will be determined so that optimum sampling and analysis procedures may be specified for network operation. The sites at State College, Pa., and Oak Ridge, Tenn., are located near calibrated watersheds and will provide comparisons between "concentration monitoring" and mass budget measurement estimates of dry deposition fluxes.

Development of Automated Instruments. A program is beginning to develop automated instruments to determine the chemical composition precipitation at the time of the precipitation event. First priority will be developing an improved pH instrument; the goal is to have a prototype available for limited field testing by the end of FY 1983.

Plans through FY 1989. The timetable for implementing various elements of the Deposition Monitoring Task Group's program is illustrated in Figure D-4.

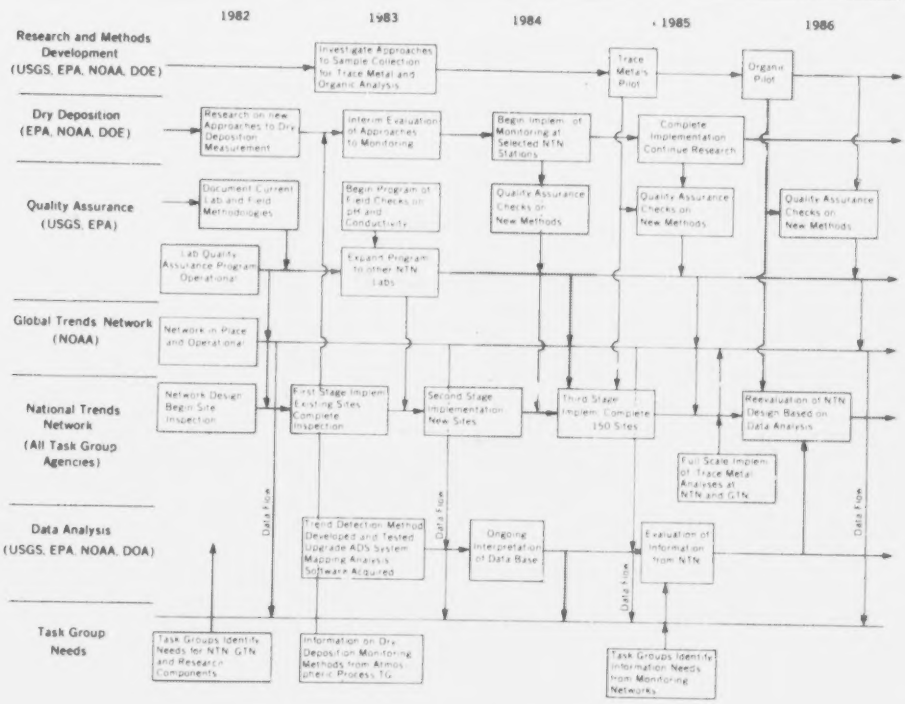
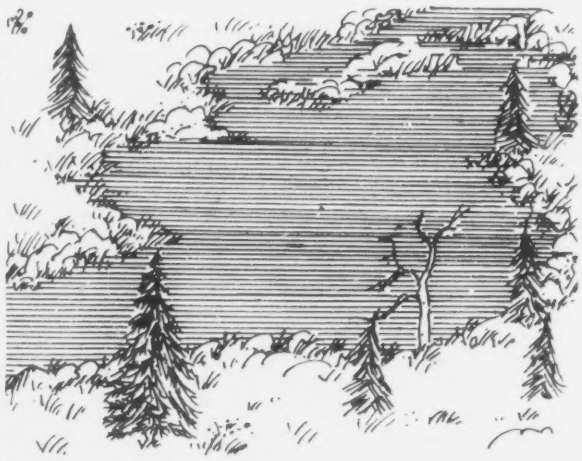

1982
1983
1984
1985
1986
Research and Methods Development (USGS, EPA, NOAA, DOE)
Investigate Approaches to Sample Collection for Trace Metal and Organic Analysis
Trace Metals Pilot
Organic Pilot
Dry Deposition (EPA, NOAA, DOE)
Research on New Approaches to Dry Deposition Measurement
Interim Evaluation of Approaches to Monitoring
Begin Implem. of Monitoring at Selected NTN Stations
Complete Implementation Continue Research
Quality Assurance (USGS, EPA)
Document Current Lab and Field Methodologies
Begin Program of Field Checks on pH and Conductivity
Quality Assurance Checks on New Methods
Quality Assurance Checks on New Methods
Quality Assurance Checks on New Methods
Lab Quality Assurance Program Operational
Expand Program to other NTN Labs
Global Trends Network (NOAA)
Network in Place and Operational
National Trends Network (All Task Group Agencies)
Network Design Begin Site Inspection
First Stage Implem. Existing Sites Complete Inspection
Second Stage Implementation New Sites
Third Stage Implem. Complete 150 Sites
Reevaluation of NTN Design Based on Data Analysis
Full Scale Implem. of Trace Metal Analyses at NTN and CTN
Data Flow
Data Flow
Data Flow
Data Flow
Data Analysis (USGS, EPA, NOAA, DOA)
Trend Detection Method Developed and Tested Upgrade ADS System Mapping Analysis Software Acquired
Ongoing Interpretation of Data Base
Evaluation of Information from NTN
Task Group Needs
Task Groups Identify Needs for NTN, CTN and Research Components
Information on Dry Deposition Monitoring Methods from Atmospheric Process TGs
Task Groups Identify Information Needs from Monitoring Networks

Figure D-4.—Proposed program implementation strategy, Deposition Monitoring

E. *Aquatic Effects*

BACKGROUND



The Aquatic Effects Task Group works to meet specific objectives that will provide information needed to assess the effects of acid deposition on aquatic ecosystems. A general summary of current scientific knowledge follows.

Acid deposition has been reported in the literature as causing both long- and short-term episodic depressions in pH, and loss in alkalinity in some lakes and streams in the United States and Canada. The total number of lakes and streams in eastern North America thought to have been acidified by acid deposition is a very small percentage of the total aquatic resource. Elevated concentrations of toxic elements such as aluminum, and biological effects, including losses in fish populations, have been reported to accompany some of these pH depressions. However, in most of the reported cases, clear relationships were not established between acid deposition and observed effects.

Current scientific knowledge does not make it possible to derive quantitative loading effects relationships for aquatic ecosystems. However, based on results of empirical studies, interpretation of long-term water quality data, and studies of sediment cores, it can be concluded that acid deposition has caused long- and short-term acidification of some low alkalinity surface waters. The purpose of the extensive research efforts in the United States and Canada is to develop such quantitative relationships as soon as possible.

Extent of acidification and sensitivity of lakes, streams, groundwaters, and wetlands of the United States. Sensitivity of surface waters to acidification is usually assessed on the basis of alkalinity. Alkalinity measures the extent to which water is buffered against chemical changes caused when acidic substances enter it. However, only a few historical records on surface water chemistry exist that can be used to assess past trends in acidification. The records available for areas exposed to acid deposition show increases in sulfate and corresponding decreases in pH for some lakes and streams. In most cases the historical data bases were not sufficiently complete to determine if a cause and effect relationship existed.

The Adirondack Mountains, located in a zone receiving highly acid deposition, comprise one of the most sensitive lake districts in the eastern United States. Fifty-two percent of 214 high elevation lakes sampled in 1975 had pH values less than 5.0. A study of 95 small, low alkalinity lakes in New England for which historical data were available showed that 64 percent had experienced significant pH decreases. Two other studies have indicated pH decreases in some lakes surveyed in Maine. On the other hand, preliminary studies suggest certain lakes in Wisconsin have shown a decrease in acidity over time.

Factors that control the susceptibility of natural waters to acidification. A complex set of factors governs the response of a body of water to acid. Geology determines the geochemical materials (thus the geochemical reactions) that acid precipitation encounters as it passes through the watershed. The hydrodynamics of the watershed determines the duration of contact of precipitation with the reaction materials. Together, these two factors govern potential chemical reactions, their extent, and their degree of occurrence. The addition of minerals other than those that produce alkalinity is critical to the aquatic ecosystem. Aluminum, in particular, appears to be mobilized by acidification of the terrestrial ecosystem and then leached into the aquatic ecosystem where it can affect fish and possibly other animals and plants.

The processes controlling sensitivity and tolerance of waters to acid loading are not yet sufficiently understood to predict the short- and long-term effects of various deposition rates on aquatic ecosystems. To generate this needed information, the Aquatic Effects Task Group is developing quantitative models that relate atmospheric influences (such as sulfate or hydrogen ion concentration of precipitation) and watershed-derived factors (such as alkalinity or sulfate concentration in lakes) to each other. Detailed mechanistic models are exploring relevant physical and chemical processes in a watershed to describe or predict changes. The Integrated Lake-Watershed Acidification Study (ILWAS) sponsored by the Electric Power Research Institute is an example. These models were developed from specific data sets, and their more general predictive value, as well as that of models yet to be developed, remains to be determined by this research objective.

Biological processes, biological populations, and aquatic communities affected by surface water acidification. Nine rivers in Nova Scotia have lost their Atlantic salmon populations while fish have survived in other rivers in the same area that have higher pH and greater alkalinity. In the Adirondack Mountains of New York, comparison of data from the 1930's with recent surveys shows that at least 180 former brook trout ponds are acidic and no longer support brook trout, although a direct association with acid deposition has not been established. The relative contribution of natural and man-made sources and changes in land use to acidification of these lakes and rivers, however, is not known.

Many species of amphibians breed in temporary pools formed by the mixture of spring rains and snowmelt. Many such pools are subject to low pH conditions. Embryonic deformities and mortalities in the yellow-spotted salamander have been observed in New York State where the acidity of meltwater pools was 1.5 pH units lower than that of nearby permanent ponds. Population densities of the bullfrog and woodfrog were less in acidic streams and ponds in Ontario versus ponds with higher pH values. These data are very limited in scope and therefore, the extent of the problem is not known.

With few exceptions, the mechanisms of effects of acidification on fish, amphibians, and other aquatic organisms are not understood. Biological changes can occur as pH drops below 6.0 and can become increasingly severe as pH declines further. Many water bodies have natural pH levels below 6.0. Aluminum and possibly other toxic elements become available in acidic waters and contribute to biological changes. These effects are currently known only qualitatively, except for the relationship between aluminum and some fish species.

Potential for acidification of surface waters, groundwaters, and wetlands to affect human health. Studies conducted in Sweden found long-term chemical changes in groundwaters from water-bearing rock formations in sandy soils near cities. Few data are available concerning changes over time in groundwater quality in the United States, although in a few cases waters from acidic wells and municipal reservoirs are associated with elevated concentrations of toxic metals leached from domestic water supply pipes. But the linkage to acid deposition, if any, remains unclear. A survey of water systems and groundwaters in the Northeast is underway, but the results are not yet available. Little is known concerning possible health implications of wetland acidification.

Mitigative (liming) techniques for restoring or protecting acidified aquatic resources of the United States. Liming is one mitigative option for temporarily protecting and possibly rehabilitating affected aquatic systems. Used in this context, liming is a generic term that indicates adding any "basic" material to surface waters, sediments, or soils to neutralize them or increase alkalinity. Liming is not considered a permanent solution to surface water acidification, but symptomatically treats the problem. Preliminary research on liming operations (primarily in Scandinavia) has indicated that some short-term protection and/or renovation of affected surface waters is possible. However, these studies are not complete and important questions remain on the long-term biological consequences of liming, its costs, application, materials and their chemical reactions, impact of episodic events, and the problems of resource management (e.g., restocking programs).

In field tests in North America and Sweden powdered limestone appears to be the most economical and effective reagent. However, it has disadvantages, such as precipitation of aluminum on lake bottoms with possible resolubilization during highly acidic episodes (such as spring snowmelt). Liming of streams has also been attempted, but with generally unsatisfactory results. Permanent limestone structures are ineffective because of biological fouling, and lime slurries are quickly dissipated, requiring continuous and expensive maintenance. The benefits and costs of liming and other actions for mitigating acidification and associated biological conditions in the United States are under study.

RESEARCH GOALS AND ACTIVITIES

The end point of the National Program's aquatic effects research is to quantify the effects of acid deposition on surface and groundwaters and wetlands of the United States, to describe how these effects can be corrected at the site of the problem, and to determine the ecological consequences of the remedial methods.

Research will also provide information needed to evaluate the current effects of acidification on freshwater chemistry; the likely sites of future water quality changes if acid deposition rates remain the same, increase, or decrease; and the extent and importance of changes in the ecosystem's plant and animal life associated with changes in the chemistry of lakes, streams, and wetlands.

The aquatic effects research focuses on five major objectives:

1. Quantify the extent of "sensitive" or "acidified" lakes, streams, groundwaters, and wetlands of the United States.

2. Identify and quantify the factors that control tolerance, sensitivity, and susceptibility of surface and groundwaters to acid deposition and determine how these factors can be used to predict future aquatic acidification under different loading rates.

3. Determine the relationships between surface water acidification and biological processes, biological populations, and aquatic communities and how these relationships can be used to predict future ecological effects.

4. Determine the potential for acidification of surface waters, groundwaters, and wetlands to affect human health.

5. Develop mitigative (liming) techniques for restoring or protecting acidified lakes, streams, and groundwaters of the United States.

Throughout FY 1982 the Aquatic Effects Task Group reviewed all current and planned aquatic effects research projects to (1) evaluate the relative overall priority of the research; (2) clearly define specific project objectives and determine their relevance to the research and assessment objectives of the National Program; (3) identify research deliverables in specific terms, and ascertain exactly how and when those deliverables will be available; and (4) assure that all researchers use compatible laboratory and

field methodologies, or that deviations from a standard methodology are made for valid reasons.

The Aquatic Effects Task Group held a partial research peer review in February 1982. A Federal effects research review a year later (in February 1983) (1) allowed the National Program's research to be reviewed in a comprehensive manner rather than as a series of discrete agency projects; (2) assessed the quality of the science; (3) provided a forum for interaction by field level scientists; and (4) evaluated the progress of the various research approaches. Throughout 1983 the Aquatic Effects Task Group will continue to critically evaluate all research to be sure that each project contributes to the identified objectives, milestones, and deliverables.

PROGRAM ACCOMPLISHMENTS—FY 1982

• **Accelerated assessment of the extent of the problem of acidification of lakes, streams, and groundwaters of the United States.** This multiagency effort comprises an assessment program to bring relevant existing water quality data together on a national basis for use in assessing the extent of sensitivity to acidification. In FY 1982 this effort was accelerated to provide results in 1983 and 1984. Specific objectives are:

1. Identify and map the extent and degree of sensitivity to acidification of lakes and streams in the United States.
2. Determine trends in the acidification of surface waters.
3. Provide a verified, quality assured data base for available methodology to predict surface water acidification based on given deposition rates. The data base will also help correlate geographic patterns of surface water sensitivity and acidification with spatial patterns of climatic, geologic, physiographic, edaphic, and land use factors.

In FY 1982 various individual projects of the National Program included a national inventory of existing water quality data applicable to assessment of sensitivity and acidification of surface and groundwaters, supplementary field surveys in the southern Appalachians, data stored in the Acidification Chemistry Information Database (ACID), and a national map indicating general patterns of surface water alkalinity (see Fig. E-1 insert on back cover).

• **The Aquatic Effects Task Group began a study to identify, quantify, and predict the factors that control the susceptibility of natural waters to acidification.** This study will mold numerous research projects into a nationwide source that will lead to a predictive modeling capability. The Task Group has begun a thorough review of the various models for predicting acidification. One or more of these approaches (or others to be developed) will be used by the National Program to predict future acidification of surface waters based on different acid loading rates for an FY 1985 assessment.

• **A major report has identified the needs of aquatic effects research.** This report reviews acid deposition phenomena in aquatic systems, develops a framework for pulling together existing information, and then evaluates and refines a preliminary model to estimate effects of acid deposition on aquatic resources.

• **A preliminary survey of New England surface and groundwater supplies was completed.** Preliminary results showed that, in general, the water supplies in the New York Adirondack region and in the New England States are quite corrosive. The most notable exception to this was the water supplies of Connecticut, which appear to be less corrosive than those in the other States. With laboratory analysis nearly completed, it appears that the maximum contaminant level for lead is seldom exceeded. Elevated concentrations have occasionally been observed, especially in standing waters. This study is being conducted to ascertain whether acid deposition might affect municipal water supplies.

• **The Aquatic Effects Task Group began a long-term program to monitor the chemistry and plant and animal life of surface waters in key sections of the Nation.** By quantifying current effects of a range of acid loading rates, the long-term surface water monitoring program is providing information needed to formulate recommendations for acceptable loading rates of acid materials from the atmosphere.

Aquatic Effects Task Group agencies cooperated in developing a standardized sampling/analysis protocol for chemical survey and monitoring of surface waters. This protocol will be completed in early FY 1983.

Sites for long-term monitoring of lakes and streams have been identified. Close coordination and cooperation among the agencies will result in an effective, compatible network. Annual reports from each site will be combined into an overall annual summary report, which will be submitted to the Task Force.

• **A number of new biological effects studies started in FY 1982.** Three different approaches (field, laboratory, and population modeling) are being used to identify and quantify the effects of lake and stream acidification on aquatic life. Studies emphasize identifying the operating stress mechanism (a toxicant and its mode of effect) and its effects on species of economic and recreational use, particularly fish and waterfowl.

Critical research issues to resolve before assessments can be made are (a) the distribution of species in sensitive waters in relation to current or future acidification; (b) biological effects of surge versus continuous acid loading; (c) individual and population effects of various levels of pH, aluminum, and heavy metals on fish and other aquatic life; and (d) sensitivity of key aquatic species, particularly fish, at various life stages to sublethal effects of low pH. The most intense work will center on areas across the Nation currently receiving acid deposition whose surface waters are highly sensitive to acidification (Northeast, upper Midwest, Appalachian Mountains, sections of the Rocky Mountains, the Cascades, and Sierra Nevada Mountains).

• **An evaluation of mitigation strategies for restoring or protecting lakes, streams, and watersheds has progressed.** An in-depth report by the Aquatic Effects Task Group summarizes up-to-date available information on liming; lists State, Federal, and private sector contacts on liming projects; and digests liming projects in the northeastern and midwestern States. Current research projects and research in Scandinavia, Canada, and the United States are included.

The Task Force organized and conducted a week-long international workshop on liming. The Aquatic Effects

Task Group developed a research strategy and set priorities for field and laboratory projects addressing liming materials and application techniques and the biological and chemical changes associated with liming.

- Laboratory and field tests are assaying the biological and chemical consequences of liming and other mitigative treatments of acidified and sensitive lake and stream systems. This project concerns the development and testing of management procedures and strategies by which lakes and streams can be at least temporarily protected from the harmful effects of acid deposition, acidification, and associated metal/ion toxicity. These tests, involving both laboratory and field investigations, include surface waters where fish populations have either been reduced by acidification or are under stress because of acidification. Another laboratory and field project was begun on placing limestone gravel in spring upwellings as a possible mitigation measure in acidic brook trout lakes.

FUTURE ACTIVITIES

Major FY 1983 to 1985 priorities of the National Program's aquatic effects research are to (1) make substantial progress in assessing the extent of the problem and place a comprehensive long-term surface water monitoring program on line; (2) develop a better understanding of the geochemical linkage between terrestrial and aquatic systems; (3) begin biological effects studies that will result in regional predictions of future acidification; (4) complete drinking water surveys of the eastern United States; and (5) begin field tests to evaluate liming options.

More specifically, highlights of expected FY 1983 accomplishments include:

Regional maps of sensitive waters of the United States. Preliminary maps developed in FY 1982 are not of sufficient detail or accuracy to allow for national assessments. Detailed regional maps developed in FY 1983 and 1984 will provide much greater detail and be based on more data.

Preliminary report on quantification of the extent and scope of surface waters at risk in the United States. Although national and regional maps are critical to the assessment of surface waters at risk, a quantitative and statistical analysis of all relevant data from the national survey (alkalinity, pH, sulfate, and others) will provide an inventory of acidified waters and simultaneous analysis of watershed data, water chemistry, and atmospheric deposition data.

Preliminary report on analysis of causative factors for alkalinity, pH decreases, and pH increases. Appropriate data from the national survey will help analyze historical trends in water chemistry and whether they can be related to atmospheric deposition.

Report on historical trends in surface water acidification from lake sediment coring studies. An alternative approach to the detection and analysis of historical trends in lake water chemistry is through the study of lake sediment cores. Core samples yield independent records of past chemistry and biology in the lake. These findings can then be compared with trends deduced from historical analyses of water chemistry.

Complete upper Midwest synoptic surveys of water quality and related plant and animal life. Studies made over the past 4 years constitute an excellent data base for comparison with future conditions in that part of the United States most densely populated with lakes.

Interim report on stream and reservoir survey of the southern Appalachians. Low alkalinity, sensitive surface waters in the southern Appalachians consist of streams and reservoirs; natural lakes are essentially nonexistent. Both short-term surveys and long-term monitoring will ascertain the distribution and numbers of these sensitive waters and their response to acidifying forces.

Final report of sensitivity to acid deposition of aquatic resources in the Rocky Mountain National Park. This will assess the extent of the problem in a pristine region and the consequences of environmental degradation.

Report on trends at active lake and stream monitoring stations. A multiagency effort is establishing long-term study sites on strategically located lakes and streams across the United States. Regular sampling of lakes, streams, and precipitation will provide needed information on the response of sensitive surface waters in different regions to atmospheric loading.

Report on methodology and data base needed to predict biological (particularly fish) resources at risk. This methodology will provide for an assessment of the effects of increased acidification on fishery resources both regionally and nationally.

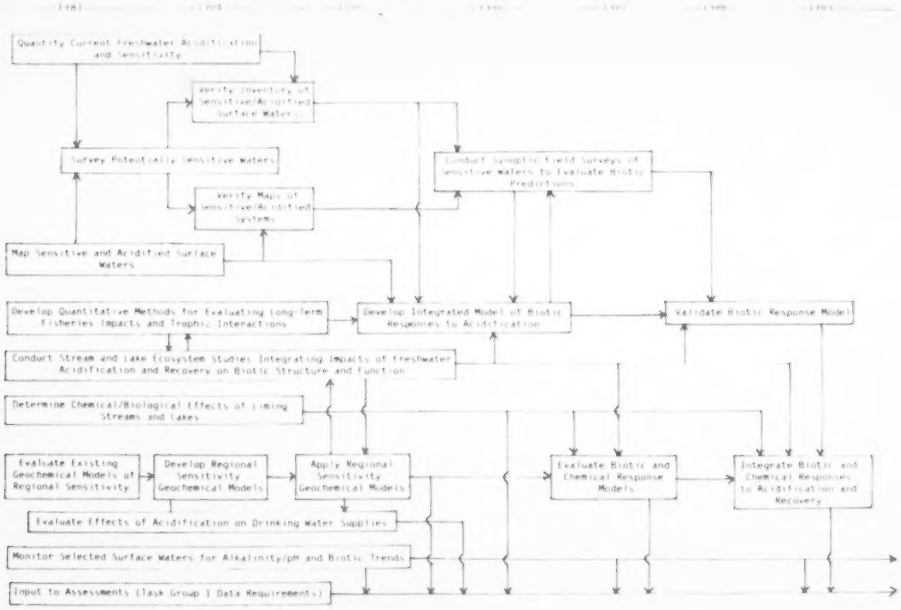

Quantify Current Freshwater Acidification and Sensitivity
Verify Inventory of Sensitive/Acidified Surface Waters
Survey Potentially Sensitive Waters
Conduct Synoptic Field Surveys of Sensitive Waters to Evaluate Biotic Predictions
Verify Maps of Sensitive/Acidified Systems
Map Sensitive and Acidified Surface Waters
Develop Quantitative Methods for Evaluating Long-Term Fisheries Impacts and Trophic Interactions
Develop Integrated Model of Biotic Responses to Acidification
Validate Biotic Response Model
Conduct Stream and Lake Ecosystem Studies Integrating Impacts of Freshwater Acidification and Recovery on Biotic Structure and Function
Determine Chemical/Biological Effects of Liming Streams and Lakes
Evaluate Existing Geochemical Models of Regional Sensitivity
Develop Regional Sensitivity Geochemical Models
Apply Regional Sensitivity Geochemical Models
Evaluate Biotic and Chemical Response Models
Integrate Biotic and Chemical Responses to Acidification and Recovery
Evaluate Effects of Acidification on Drinking Water Supplies
Monitor Selected Surface Waters for Alkalinity/pH and Biotic Trends
Input to Assessments (Task Group I Data Requirements)

Figure E 2 – Proposed program implementation strategy, Aquatic Effects

F. *Terrestrial Effects*

BACKGROUND

Only recently have efforts been made to establish the mechanisms by which atmospheric acid is transferred to aquatic ecosystems. If acid precipitation must pass through the terrestrial ecosystem prior to entering an aquatic ecosystem, it will usually be strongly influenced by the chemical nature of the vegetation, soil, and bedrock.

Aluminum movement from terrestrial systems has recently become a primary concern. Aquatic biologists have documented the effects of aluminum on fish and other aquatic organisms. It has been shown that aluminum does move into the aquatic system at a steady rate, increasing to high levels during spring snowmelt and heavy rainfall periods, and that the aluminum is coming from the terrestrial system, not the atmosphere. The availability of aluminum in the soil solution has direct implications for tree growth, and thus forest productivity. The tolerance of crops to soil-water aluminum is well known, but the toxicity data for forest trees and range plants are not available.

Terrestrial effects research encompasses soils, watersheds, forests and range plants, and crops. A number of uncertainties shroud the relationship of acid rain to the dynamics of these systems. Whether acid deposition is wet or dry, the interaction of ozone and sulfur dioxide and the life stage of the plant can modify plant response to acid deposition. That such factors vary by region adds to the uncertainty.

Other natural stress factors, such as insects and disease-causing agents, are capable of limiting forest and range productivity in the absence of acid deposition. Even if acid deposition should not prove to directly harm forest and range vegetation, it remains important to find out if it impairs the ecosystem's ability to rebound from common natural stresses—insects, disease, and drought among others—and stresses from man-made pollutants. Studies must consider how variations in precipitation chemistry may affect plant response.

While there is general agreement that unmanaged soils in forested and grassland areas in humid regions may be sensitive to acidification from acid precipitation, there is no indication to date that the soils have become acid because of it or that forest production is being affected.

Soil acidification by natural processes may be more significant than soil acidification by acid deposition. The rate and extent to which such acidification is occurring in nature, and the effects (if any) of acid rain on soil microbial processes are little understood.

The most consistent conclusion to be drawn from agricultural research at all scales and with all species has been "no effect" at current average ambient pH levels (pH 4.0-4.2). A few studies, including some with field corn and soybeans, have reported negative impacts of acid deposition, while still others showed a positive growth response. The economic importance of corn and soybeans and their location within the region most heavily exposed to acid rain and ozone warrants further studies to understand the nature of their response.

RESEARCH GOALS AND ACTIVITIES

The terrestrial effects research of the National Program includes work on the direct or indirect effects of acid deposition on three major components: forest trees and range plants, agricultural crops, and soils and watersheds. The objectives of the research are to address the following key questions:

1. What are the long-term effects of acid deposition on forest and rangeland productivity?
2. Can mathematical models be developed to estimate economic losses in major crop plants?
3. What are the relationships of acid deposition effects on soil chemistry to biological productivity?
4. What are the influences of soil weathering, leaching, and organic decomposition on ecosystem linkages among soil, water, and plant components?
5. Will acid deposition have a detrimental effect on forest and range ecology?
6. Are there threshold doses above which crop productivity will be adversely affected?
7. What are the ecosystem impacts of acid deposition, and can management options be developed to counteract them?

8. What are the physiological and biochemical processes by which plants may be changed by acid deposition?

To determine the effects of acid rain on forest productivity, their magnitude, and how they work, research is surveying trends in forest growth, determining through mechanistic studies, whether cause and effect relationships exist, and evaluating acid deposition's effect on the response of forest and range vegetation to other stress factors.

Agricultural research is focusing on major economic crops, such as corn and soybeans, and concentrating on those questions that can be most readily addressed within a 5-year time span. The emphasis is to generate realistic response functions that can be used in an assessment.

Effects on soil productivity are also being emphasized, as are mitigation measures with practical applications. Studies on soils and watershed processes are determining the changes that occur after acid deposition reaches the soil surface.

PROGRAM ACCOMPLISHMENTS—FY 1982

Forest Trees and Range Plants

Studies on forest and rangeland vegetation indicate that the effects of air pollution in general, and acid deposition in particular, are subtle and of long duration, and may alter the dynamics of plant community growth and succession. To provide the information for both economic and ecological assessments, studies emphasize the mechanisms that affect growth or mortality.

- **Tree ring sampling initiated.** Establishing regional tree growth rate changes is an essential assessment step. Changes in diameter growth can be easily converted to estimates of volume, which can subsequently be translated into economic terms. Testing is also underway to determine if trees are taking up unusually large quantities of metals; this is considered an indicator of anthropogenic activity. To assess growth rate changes over time, sampling has begun of tree ring cores in over 600 trees in 70 forest stands in 14 eastern States. Those States range from Maine to North Carolina, and extend west as far as Arkansas and Missouri.
- **Study of red spruce dieback started.** Because of both direct economic and aesthetic considerations, there is concern about red spruce dieback in the eastern United States, especially at high elevations. Dieback was generally preceded by an abrupt reduction in growth which began in 1960. At that time, a severe drought occurred in this area and may have been a triggering agent. Previous tree ring chronologies indicate fairly rapid recovery in the same systems from earlier droughts. Recovery from the current drought appears to be quite slow or nonexistent, possibly because of interactions with other stresses, both natural and anthropogenic.
- **Pine seedlings vary in their response to acid rain.** Some pine seedlings showed greatest growth response below pH 4.0, perhaps because of the nutrient effects of sulfur or nitrogen. Since southern pine forests are frequently fertilized, significant inputs of essential nutrients from atmospheric sources could substantially reduce fertilization costs.
- **Some loblolly pine seedlings show resistance to disease following acid rain exposure.** Fusiform rust is estimated to produce losses in yield in excess of $25 million per year in southeastern pine forests. Research suggests acid rain may increase rust resistance in pine. In response to simulated acid rain, some seedlings showed increased rust resistance traits, others decreases in fusiform rust galls.

Agricultural Crops

Several developments in methodology for studying acid rain effects on agricultural crops have been tested during FY 1982 that promise to improve evaluation. The ambient rainfall exclusion technique was perfected, a significant development. Researchers at six sites are developing such facilities for assessing crop loss. Field research with simulated rain, excluding ambient rainfall, is the state of the art in crop loss assessment relative to acid rain impacts. Significant progress in crop loss assessment is anticipated from the results of the 1983 growing season, as these key new experiments are performed.

CROP SENSITIVITY:

- **Potato study shows no apparent foliar injury from acid rain.** Potatoes are a major agricultural crop in the northeastern United States. In a field study, a commercially important potato cultivar was treated with simulated acid rain of various pH levels, similar to ambient rainfall at that site. No apparent foliar injury resulted from the treatment, even at levels as low as pH 2.7. Quality analysis of the 1982 tuber crop is in progress. This project will yield dose response data for both crop yield and crop quality. Both of these values are necessary for economic assessment of impacts to potatoes. In addition, information gained as a byproduct of the quality analyses will aid in understanding the mechanisms of effects.
- **No change observed in balance between clover and fescue.** Clover/fescue ratio is a good indicator of stress; both are important forage species that frequently are not as intensively managed as agricultural field crops. Therefore, it is important to see if soil management should be changed to maintain a clover-fescue forage system in the presence of acid rain. Data gathered through the first two harvests of this study showed no foliar necrosis or differences in yield or species composition that could be attributed to rain pH or soil management variables. However, in both species, the crop's nutritional quality appeared to decline at the lowest treatment (pH 3.5), indicating potential for long-range impacts on the nutrient and, therefore, market values of the crop.

DOSE-RESPONSE RELATIONSHIPS:

- **Soybean yields may or may not be adversely affected by acid deposition.** Studies indicate that yield reductions of sensitive cultivars were caused by decreases in number of pods at harvest, even without visible foliar injury. Some cultivars, however, are resistant. These results underline the need to develop damage functions for representative vegetation across a range of genetic and environmental variables.

• **A major field experiment established in the corn/soybean belt of Illinois** is dedicated to developing dose-response information on two varieties each of soybean and field corn using ambient rainfall exclusion and rainfall simulation techniques that permit study under undisturbed field conditions. This major experiment will significantly expand the data base on agricultural effects for the 1985 assessment.

• **Inhibition of corn yield.** A study of field corn using simulated acid rain between pH 5.6 and 3.0 showed inhibition of yield at pH 4.0 only, corroborating earlier reports with the same crop, and indicating that factors in addition to hydrogen ion are controlling the magnitude and direction of the effect. Mechanisms associated with the observed effect must be better understood before predictive assessments are possible with corn.

• **Concentration and ratios of ions.** Acid precipitation may stimulate plant growth and yield depending on the environmental conditions and the concentrations and ratios of ions in the simulant. Soybean yield varied depending on the sulfate to nitrate ratio in precipitation simulants; the response depended on pH. Since the sulfate to nitrate ratio in precipitation varies by region, this factor is an important consideration in regional assessments leading to the 1985 assessment.

• **Individual events.** The response of radishes and bushbeans to acid rain simulant was also significantly affected by the chemistry of individual events. Peak acidity events may be more important in affecting plant response than the total mean acidity during a growth season. Since current deposition monitoring does not measure peak acidity events, but rather, weekly average concentrations, research such as this may provide valuable feedback to research planning activities in the Deposition Monitoring Task Group.

Soils and Watersheds

Watersheds represent a linkage between terrestrial and aquatic systems critical to understanding changes in both forest and aquatic productivity and stability. Chronic disruption of nutrient cycling processes or the mobilization of nutrients and toxic elements in terrestrial and aquatic systems will probably cause long-term effects on both forest and aquatic productivity.

Current studies are aimed at: (1) evaluating characteristics that determine soil vulnerability in an attempt to establish criteria by which individual soil types might be categorized as to potential for either nutrient loss or toxic element mobility; (2) identifying the principal soil processes that can be altered by acid deposition, in order to understand the mechanisms controlling nutrient availability for plant growth; and (3) determining the role that vegetative cover plays in modifying acid deposition effects on both the terrestrial system and the transport of ions to aquatic systems.

SOIL VULNERABILITY:

Soils exhibit a broad range of potential for change in chemical and biological properties resulting from acid deposition. Possible effects include leaching of essential alkaline cations with subsequent decrease in soil fertility in addition to release of aluminum and heavy metals. Studies are underway to investigate a range of soils varying in their vulnerability to change.

• **Significant removal of nutrients observed in a southeastern soil.** Among the soils considered most vulnerable, a sandy soil from the southeastern United States was exposed in both field and laboratory to simulated rain as low as pH 3.0. At pH 3.7, significant amounts of calcium and potassium were removed from the top 76 cm of this sandy soil. Magnesium, calcium, ammonium, phosphorus, sulfate, and aluminum were all higher in the soil solution at the surface and decreased steadily with depth. Nitrate nitrogen increased with soil depth. Ions in soil solution were more concentrated during dry periods or periods of low rainfall. All of these factors could have an effect on soil fertility and, therefore, productivity and the economic return on investment.

• **No significant changes noted at a New Jersey site.** Soil solution and stream pH at a forested site in New Jersey were not directly related to precipitation pH, apparently because of natural production of acids by the soil. Large quantities of potentially mobile aluminum were found in the upper mineral soil horizons and forest floor; however, the sampling period did not include enough acid precipitation events to permit evaluation of the relationship between soil solution, aluminum concentrations, and precipitation pH.

• **Aluminum leaching found to vary by soil type.** A study of three contrasting northeastern forest soils showed the soils vary in their susceptibility to leaching of aluminum to acid precipitation. Two factors were suggested as regulating aluminum mobility in these soils: the form and quantity of the aluminum and the presence of mechanisms that favor the retention of otherwise mobile anions.

SOIL CONDITIONS AND PLANT GROWTH:

• **Acid rain causes varied response in nutrient formation in some soils from northeastern watersheds.** In laboratory studies with soils from three northeastern watersheds, simulated acid rain inhibited nitrogen mineralization in some soils, had no effect in other soils, and stimulated nitrogen mineralization in still other soils. Among the soil processes important to regulating nutrient availability for plant growth are those involving the mineralization of nitrogen by microorganisms. The nutrient most often limiting plant growth and forest productivity in the Northeast is nitrogen, and nearly all that nitrogen is derived from the microbial mineralization of nitrogen. Because few soils were studied, it is not possible to predict the soils or regions in which the nitrogen-supplying capacity of soils (and hence the provision of the limiting nutrients for vegetation) will be inhibited, enhanced, or unaffected by acid rain. Additional information from other sites is needed to establish regional responses for assessment purposes.

• **Acid rain increases nutrient availability at a southeastern site.** Soil properties and environmental conditions may also play an important role in microbial processes, as indicated from the results of a southeastern United States study where acid precipitation increased the rate of both nitrification and mineralization. Denitrification occurred in the acid sandy soils with low organic mat-

ter content; however, the effect of acid precipitation on denitrification is not clear at this time. Acid precipitation of pH 3.7 did not have any significant impact on soil biomass, respiration and enzymatic activity; however, the soil type may be a very important influence.

• **Sulfur accumulates in forested southeastern watershed.** It was observed that sulfate accounts for about 60 percent of the anions present in North Carolina Appalachian precipitation. This study of mineral cycling processes in a forested southeastern watershed found accumulation of elemental sulfur, because of its adsorption and transformation into nonmobile forms. Both processes are very important in regulating soil and water chemistry and acidity by retarding anion mobility and, thereby, cation nutrient loss from terrestrial systems.

WATERSHED BUFFERING CAPACITY AND RESPONSE:

• **Precipitation pH was not altered in one New England watershed, though the source of acid precipitation precursors may have changed.** As part of a long-term study of nutrient cycling budgets relating to forest management, no significant shift in precipitation pH was found at Hubbard Brook watershed in New Hampshire. These same results, however, indicate a decrease in the ratio of sulfate to nitrate, suggesting an important shift in the source of acid precipitation precursors.

• **pH of precipitation altered by forest ecosystems.** Forest ecosystems can reduce the acidity of ambient rainfall. Rainfall at pH 3.5 to 4.5 on a hardwood forest in New Hampshire was buffered to pH 5.0 by the time it reached streamflow. A West Virginia watershed with low alkalinity received rainfall at pH 4.1; below the tree canopy, it had risen to pH 4.2; in groundwater at springs, it was pH 5.0; and at the outlet of the small watershed, pH 5.9. Aspen forests growing on slightly calcareous glacial till in Minnesota also effectively buffered the impact of pH precipitation events.

However, in another Minnesota tree stand, while the pH of precipitation passing through black spruce growing on peat land increased slightly, the wetland surface peat added significantly to hydrogen ion concentration and ultimately reduced pH to the 3.5–4.5 range. Red pine growing on shallow soil over granite bedrock added a slight amount of hydrogen ion to precipitation, but the soil subtracted about the same amount. In this case, incoming precipitation is fed with little change in pH directly to streams and lakes. The mixed or variable nature of these observed responses demonstrates that variables such as canopy species, soil, and bedrock clearly must be accounted for in estimating deposition inputs to aquatic ecosystems from terrestrial ecosystems.

• **Water quality data lacking in some southeastern States.** A nationwide assessment of existing water quality data indicates that there is insufficient information on water quality variations in small streams during storms in southern Appalachia.

• **Site-specific deposition data needed.** To extrapolate site-specific data to regional effects, it is necessary to know if environmental conditions where the data are gathered are representative of the region. The extrapolated information is used in dry deposition models to quantify acid additions to the terrestrial ecosystem. Two types of currently available deposition models were evaluated for their applicability to developing watershed mass balance estimates. Although both models are state of the art, both have proved to be either inaccurate for most mass balance applications, or have inadequate characterization of major measurement parameters. This study has identified the fact that the quality and level of resolution of the models tested will need to be improved to support the level of detail needed for proposed ecological and economic assessments.

FUTURE ACTIVITIES

In the near future, research findings will accelerate as results of multiyear research activities are summarized. Among expected results between 1983 and 1985 are thresholds of damage to agricultural crops and dose-response relationships, specifically for corn and soybeans. This information will be used in developing models to estimate economic impact.

In forest ecosystems, tree ring studies will indicate the potential for determining growth changes related to acid deposition, as well as abnormal accumulation of metallic ions. There will be a preliminary report of soil nutrient budgets responsive to acid deposition. Changes of forest productivity correlated with acid deposition effects also will be summarized for major forest types. The role that various types and densities of vegetative cover play in modifying acid deposition effects on both the terrestrial system and the transport of ions to aquatic systems is being studied. Work beyond 1985 will concentrate on determining whether acid rain has predisposed trees to moisture stress.

Understanding the effects of acid deposition on the terrestrial ecosystem will be greatly improved by soil studies of aluminum and other metals, their mobility, and their uptake by both forest and agricultural crops, coupled with improved understanding of form and amount of acidic materials delivered to the forest floor. A report on the significance of hydrogen ion generated in soils will be prepared in 1985 as will reports on aluminum mobilization and on mineralization processes affected by soil acidification.

From 1986 to 1988 efforts will be accelerated to categorize soil types as to nutrient loss or toxic element mobility, as will studies to determine which soil processes controlling nutrient availability for plant growth will be altered by acidic fluxes. By 1988 these studies will begin to provide correlations that can be used to improve models. This will greatly aid the understanding of the influence of vegetative cover and soil behavior on water quality eventually reaching streams and lakes.

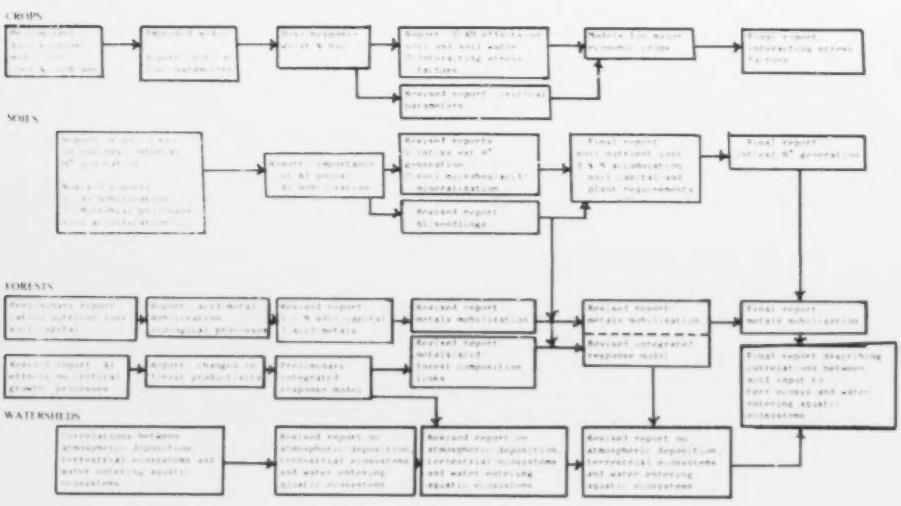
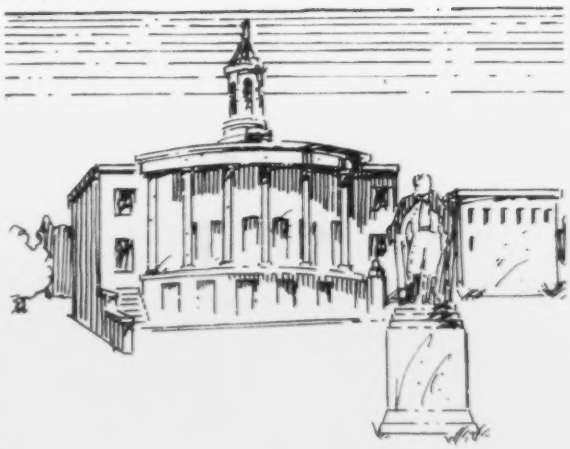


Figure F.1. Proposed program implementation strategy, Terrestrial Effects.

G. *Effects on Materials*

BACKGROUND



Experience over the last century in both European and North American cities has shown that air pollutants accelerate the deterioration of materials. However, several factors complicate quantitative assessments of the impact of atmospheric pollutants, including acid precipitation, on conventional building materials. Building materials degrade to some extent with time, even in pristine environments, so it is important to differentiate between normal weathering and deterioration accelerated by air pollution.

Distinguishing between the relative amounts of damage caused by specific pollutants and their transformations in contact with various surfaces is even more elusive. The role played by acid precipitation is still a major unknown. This is especially true for the acid precipitation stemming from long-range transport of air pollutants.

The development of quantitative cause and effect relationships depends on the capability to correlate two independent data sets – materials deterioration and pollution monitoring.

Past attempts to estimate the materials damage costs related to pollutants are of questionable value, because:

1. The effects caused by various pollutants cannot yet be reliably separated from each other and from deterioration caused by natural agents such as moisture and temperature.
2. Estimates of the quantities of materials exposed to acidification are lacking.
3. The way society chooses among use, repair, and replacement of materials susceptible to air pollution damage is unclear.

Materials at risk are statuary and structures of cultural value as well as commonly used construction materials. These include man-made materials such as metals and alloys, paint, plastics, cement and concrete, masonry, roofing and siding materials, and natural materials such as marble, limestone, granite, calcareous sandstone, and other mineral substances. Inventories of the location and amounts of sensitive materials present in the geographic areas of concern do not exist. Such inventories must be completed if the economic effects of air pollution damage are to be assessed and comprehensive cost estimates developed.

Chemical corrosion processes deteriorate calcareous building materials and cause metals, particularly zinc and copper, to lose their protective oxide coverings. Over the years, a considerable amount of industrial research has evaluated the relative susceptibilities of materials and developed more resistant ones. However, little has been done to quantify the susceptibility as a function of pollutant concentrations or other exposure variables such as humidity, temperature, wind direction and velocity, precipitation quality and frequency, or the concentration of airborne salt particles.

Climatic and meteorological conditions are important unknowns with respect to materials damage, although some information is available. For example, it is known that high humidity increases the rate of attack of sulfur dioxide on zinc, and that local variations in temperature also can be expected to influence damage rates. Wind velocity and direction can affect the deposition velocity and also enhance surface abrasion by large airborne particles. An important role in materials damage also has been ascribed to special events such as dews, frosts, and fog in combination with dry acid deposition.

The principal air pollutants causing materials damage are sulfur dioxide, nitrogen oxides, ozone, and particulate matter. In addition to attacking surfaces directly, the first two are precursors of particulate sulfates and nitrates that can travel long distances and, as acid deposition, create effects many miles from the original emission sources. However, since pollutants are rarely encountered singly and are thought to act synergistically, they must be examined in combination and in different proportions corresponding to actual conditions. Information regarding the potentially important effects of ammonia and chlorine is also unavailable.

RESEARCH GOALS AND ACTIVITIES

Assessing damages to materials and cultural resources by acid deposition was a major new FY 1982 initiative of the National Acid Precipitation Assessment Program. The Task Force is planning and beginning several ac-

tivities to better understand and quantify the role of acid deposition in degrading various materials and cultural resources. The present state of the art of acid deposition materials damage modeling and related economic effects modeling is primitive and the required data bases are very poorly developed. The Materials Effects Task Group has designed a research strategy that will provide basic understanding of damage mechanisms from which effective protective measures for historic and cultural structures may be developed and from which more accurate economic estimates can be made.

The Materials Effects Task Group's research strategy addresses four key questions:

1. What portion of materials damage can be ascribed to acid deposition?
2. Which materials are susceptible to which specific air pollutants and to what degree?
3. What is the geographic distribution of susceptible materials?
4. What are the specific rates of damage or amounts of damage in economic terms?

PROGRAM ACCOMPLISHMENTS—FY 1982

In its first year, the Materials Effects Task Group concentrated on developing interagency coordination and cooperative study, and implementing new projects. Efforts during FY 1982 concentrated on (1) expanding field studies of materials deterioration; (2) intensifying efforts to look retrospectively at materials damage on marble tombstones, exposed bronze markers, and materials performance data from industry-sponsored research; (3) selecting and applying methods to measure air quality for the purposes of materials damage research; and (4) developing methods to inventory common construction materials as well as historic structures and monuments.

Research initiatives in FY 1982 included:

- **Establishing new field sites.** Two additional field sites were established at Washington, D.C., and Huntington Forest, N.Y. Unlike the first two sites at Chester, N.J., and Research Triangle Institute, N.C., these new locations did not have full environmental monitoring instrumentation and therefore various supplemental monitors had to be installed. Atmospheric corrosion testing, coordinated with simultaneous meteorological, acid deposition, and air quality measurements, is being conducted at these four field sites to quantitatively correlate specific pollutants present in acid deposition with materials damage. The corrosion responses of seven metals are being evaluated over exposure cycles ranging from 1 month to 3 years. The result will be dose-response functions for these metals.

A related problem now under investigation is the proper method of sheltering a set of samples from wet deposition. This work has produced a prototype movable cover activated by a rain sensor. This movable cover is now being evaluated for possible installation at Bureau of Mines sites. This will permit wet and dry deposition effects to be studied separately.

- **Calibrating static monitors for sulfur dioxide and nitrogen oxides.** These simple devices appear to promise a much cheaper method of characterizing air pollution levels at materials effects sites. However, the literature contains little concerning their performance under varying conditions of wind, humidity, temperature, and combined pollutants. Therefore, experiments have begun to study the response of these monitors, beginning with a 1-month field exposure during June 1982, as part of a major project on dry deposition conducted at Champaign-Urbana, Ill., for the National Program.

In this project, a variety of materials were exposed with the static monitors to compare their relative rates of sulfate and nitrate uptake. Extensive meteorological data were collected simultaneously. In addition, a flow chamber for calibrating the static monitors under controlled conditions was designed and built at Research Triangle Institute. Actual operation of the flow chamber will begin in early FY 1983. The availability of properly calibrated static monitors may ultimately permit operating materials exposure sites without elaborate air quality monitoring equipment.

- **Estimating damage functions from industry site data.** Much of the basic data are derived from commercial material test sites. The National Bureau of Standards (NBS) is collecting existing corrosion and material performance data. Methods are being developed to reconstruct past pollutant deposition levels for the regions and periods of commercially-sponsored materials exposure testing. Available material performance data have been identified and will be compiled in FY 1983, including historical pollutant levels and meteorological data. Correlation of these data sets will provide rough, interim estimates of material deterioration effects attributable to acid deposition until more definitive damage functions can be developed by field exposure corrosion testing.

- **Estimating bronze deterioration rates.** A project to estimate bronze deterioration rates resulting from exposure was begun in FY 1982. Bronze markers such as U.S. Geological Survey surveyors' benchmarks and National Historic Landmark plaques will be studied. In FY 1982, NBS tested and refined a mylar replica method to measure the rate of corrosion in the field. Bronze durability as a function of deposition levels will be estimated.

- **Estimating marble deterioration rates from tombstones.** Study of the differential rate of marble weathering, as measured on Veterans Administration tombstones, has produced a data base covering 3,500 individual stones. The data are now being analyzed and correlated against environmental factors, including air pollution levels. The final report is due in FY 1983. In addition, a report has been completed on maintenance practices in NPS cemeteries. The output of the tombstones study will be a set of correlations between marble deterioration rates and spatial and temporal variables, including retrospective estimates of pollution levels.

- **Pollution archaeology.** Success of the retrospective studies of materials damage requires estimates of past pollution levels. Since air pollution was not usually monitored prior to 1960, these levels must be inferred from other data sources. Two such efforts were begun in FY 1982. One approach involves calculating air pollution levels from historical records of fuel consumption. The estimates will use primarily local sources of data covering sources such as home heating, industries, and vehicles, including coal-fired locomotives. In addition, NBS began to develop a method to measure levels of at-

mospheric sulfur deposited on painted surfaces and trapped between repaintings. If measuring sulfur levels in lead paint layers proves feasible, a field study collecting paint samples from dated structures nationwide will be started in FY 1983 to verify sulfur deposition levels calculated from emissions inventories, and to refine regional scale estimates of deposition levels.

• **Prototype *in situ* deterioration monitoring.** Diagnosis of actual cases of materials deterioration is the practical application of results of field, lab, and retrospective research. As a prototype, deterioration of historic structures at Mesa Verde National Park, Colo., will be measured in parallel with environmental parameters. National Trends Network parameters will be supplemented by measures of gaseous concentrations, particulate composition, and temperature and moisture cycles. Field measurement techniques perfected at the prototype site will be available for general application nationwide.

• **Documenting protective treatments.** Baseline documentation of effective protective treatments for masonry monuments and buildings was undertaken for eight structures in FY 1982. Structures were chosen based on several criteria: masonry material, treatment material, and location of structure. Materials include marble with and without epoxy consolidants, sandstone with epoxy consolidants, cleaned brick, and terra cotta. Locations include New York, N.Y., Portland, Ore., Minneapolis, Minn., Chicago, Ill., San Francisco, Calif., New Orleans, La., and Boston, Mass. The results will be used to assess existing techniques for protecting such structures against environmental decay.

FUTURE ACTIVITIES

In FY 1983, work will continue toward producing interim damage functions for a 1985 assessment. Tombstone data, bronze marker data, and material performance data from industry will be correlated with reconstructed pollution histories. Several laboratories will begin to adapt advanced technology to increase the sensitivity of damage measurements; this will shorten the time frame for determining damage function. Work will be undertaken to characterize the mineralogy of major American stone sources and to predict their susceptibility to attack by acid deposition.

Chamber studies to investigate the role of moisture in acid deposition upon material surfaces and the effectiveness of protective treatments on statuary materials will begin in FY 1983. Additional materials – marbles, bronzes, and selected coating materials – will be added to the field exposure program.

Another initiative will develop data on the distribution of construction materials as related to population, history, availability of materials, and land use. This data will be incorporated into a land use classification that will produce an inventory of common construction materials.

The Materials Effects Task Group presently plans the following FY 1983 outputs:

Tombstone study results – a report of marble damage rates.

Bibliography of Effects of Acid Deposition on Corrosion and Deterioration of Materials, 1980-1983 – a literature review covering existing knowledge of materials damage.

Description of field exposure sites and methods – a report setting forth for peer review the field exposure program.

Specifications for measuring damage to cultural resources – a document establishing the basis for investigating damage at cultural sites from acid deposition and other causes.

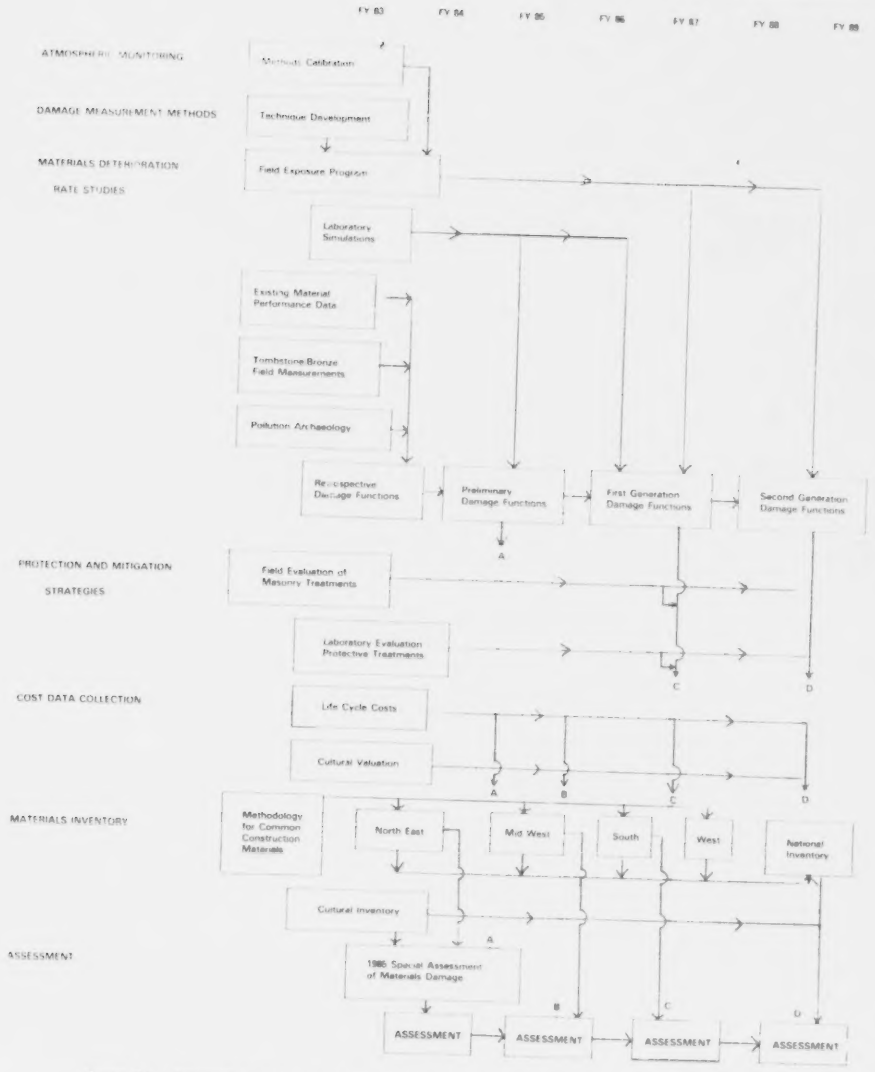


Figure G1. Proposed program implementation strategy. Effects on Materials and Cultural Resources.

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H. Control Technologies

BACKGROUND

The relationship between man-made pollutants and acid deposition is not fully understood, but three pollutants have been linked to acid deposition. Sulfur oxides and nitrogen oxides have been analytically identified in acidic deposits with the ratio of each ranging from minor to major. Oxidants also have been highly correlated with the occurrence of acid precipitation and are probably involved in the atmospheric transformation of the gaseous oxides to acidic nitrates to sulfates that are deposited.

Emissions from fossil fuel power plants have been the focus of attention for possibly controlling the release of acid precursors because nationally this source produces a major share of the sulfur dioxide and a sizeable portion of the nitrogen oxides. However, other activities such as smelting, industrial combustion and processes, transportation, and space heating can all contribute significantly to the atmospheric loading of sulfur and nitrogen acid precursors and oxidants. The following discussion primarily addresses the utility source because our knowledge about the importance of other sources is less complete. All may contribute significantly (locally and/or regionally) to the formation of acid deposition, but their specific roles are still poorly understood.

Table H-1 summarizes the major man-made acid precipitation precursor pollutants, the major industrial sources, applicable controls, and explains their operation. For sulfur dioxide and nitrogen oxides, generally available controls are either expensive or inefficient. However, important technologies being developed, such as low nitrogen oxide burners and limestone injection multistage burners (LIMB), could someday provide more cost-effective control of acid precipitation precursors. For the directly emitted acidic compounds, very little information is available on the effectiveness of conventional control technology.

Table H-2 focuses on coal-fired power plants, major emitters of sulfur dioxide and nitrogen oxides pollutants. The table summarizes technologies that can be retrofitted to the sources of these man-made pollutants by performance, commercial status, costs, applicability, and important problems.

Flue gas desulfurization (FGD) technology is capable of highly efficient sulfur dioxide removal and has been commercially available for over 10 years. However, its capital costs are quite high; such systems would cost $60 to $90 million for a typical 300 MW coal-fired utility boiler. Also, widespread use of such systems would generate large quantities of wet solid waste product that must be disposed of in an environmentally acceptable manner.

Physical coal cleaning, although significantly less expensive than FGD, can decrease sulfur dioxide emissions only modestly since chemically-bound sulfur cannot be removed with current coal cleaning technology.

While switching to low sulfur fuel can achieve significant reductions, its capital costs can also be quite high — particularly when major boiler derating is needed to accommodate the lower fuel heat content of western low-sulfur subbituminous coals or particulate systems that require upgrading because of different coal effluent characteristics. Costs for this method also increase if locally mined coal can no longer be used.

Among the emerging control technologies, low nitrogen oxide burners may be particularly effective in controlling the oxides of nitrogen and may offer relatively inexpensive capital and operating costs, but achieve no sulfur dioxide control.

LIMB technology appears capable of moderate to high removals of both pollutants at less than 20 percent of FGD costs. LIMB technology is now in the prototype stage, but a demonstration may provide sufficient data by 1985 to permit industry to begin to evaluate its potential. Not enough information is now available to determine if target removal efficiencies can be met by LIMB without introducing boiler operating problems.

TASK GROUP ROLE

The Control Technology Task Group is tracking all relevant control hardware development efforts, and provides the appropriate agencies with guidance related to the National Program's objectives and requirements.

The interagency budget for the National Program does not include Federal funds for control technology hardware development because control technology activities are

conducted under preexisting programs at EPA, DOE, and TVA. The various reasons for controlling the primary man-made acid precursors, sulfur dioxide and oxides of nitrogen, make it difficult to directly assign the cost of such emission control hardware work to such different concerns as health, visibility, and acid rain. The National Program is actively coordinating with the relevant ongoing Federal control technology efforts to ensure that the concerns relative to acid deposition are addressed. A summary follows of the relevant Federal research and development activities in the existing control programs.

Technology	Agency	FY 82 ($ in millions)
SO_2 Control[1]	EPA	2.0
SO_2 Control	TVA	2.6
NO_x Control[2]	EPA	3.9
LIMB[3] Low NO_x Burner	EPA	7.2
Simultaneous NO_x SO_2	DOE	6.2
Total		21.9

[1] SO_2 = sulfur dioxide
[2] NO_x = oxides of nitrogen
[3] LIMB = limestone injection multistage burner

In addition to these Federal efforts, it is anticipated that the private sector will play a major role in bringing promising new technologies to the commercialization stage.

TECHNOLOGY DEVELOPMENTS—FY 1982

During FY 1982, the sulfur dioxide and oxides of nitrogen control technology activities funded by EPA, DOE, and TVA made significant progress in developing improved methods for controlling these emissions. The Control Technologies Task Group has identified the following results as potentially relevant to addressing the acid deposition issue:

- **Successful initial tests of the Limestone Injection Multistage Burner (see Figures H-1 and H-2).** Encouraging initial results indicate the potential for LIMB as a low-cost retrofittable combination nitrogen oxides and sulfur dioxide control technology. The LIMB technique uses sorbent injection of limestone during combustion with low nitrogen oxides burners. This developing technology may be able to reduce these pollutants' emissions from utility boilers by over 50 percent. (EPA)

- **Development of burner systems that could reduce nitrogen oxides emissions by 75 percent.** Advanced burner systems for wall-fired and tangential-fired boilers have been tested that decrease nitrogen oxides emissions about 75 percent. Since coal-fired utility boilers generate 40 percent of the total nitrogen oxides from stationary sources, these new burner systems should have an important influence on future emissions from utility boilers. (EPA)

- **Improved sulfur dioxide wet scrubbing techniques.** Wet limestone and lime flue gas desulfurization process technologies have been improved by increasing sulfur dioxide removal efficiency and reagent utilization, upgrading reliability, and improving waste product disposal. Innovations include adipic acid, magnesium, and thiosulfate additives; forced oxidation; and scale prevention in scrubbers and mist eliminators. These techniques improve sulfur dioxide removal efficiency and process reliability to over 90 percent while reducing costs. (EPA, TVA and DOE)

- **Evaluation of dry scrubbing process.** The researchers evaluated the effectiveness of spray dryer flue gas desulfurization systems using lime, limestone, nahcolite, and trona, and investigated the relative merits of fabric filters and electrostatic precipitators for collecting fly ash and sorbent material. This technology provides a new and lower-cost option than wet scrubbing processes for controlling sulfur dioxide from certain utility boilers. (EPA and DOE)

- **Assembled coal data bases.** Data bases on the quantity, sulfur content, heating value, ash content, and desulfurization potential of U.S. coals were assembled. These permit evaluation of the amounts of coal from each State that can be used with different control technologies for compliance with differing sulfur dioxide emissions control regulations or strategies. (EPA and DOE)

- **Evaluated advanced simultaneous sulfur dioxide/nitrogen oxides flue gas treatment processes.** Progress was made in quantifying the cost and performance relationships, and identifying other promising emerging processes, such as electron beam irradiation, for further research. From this research may well evolve the next generation of air pollution control technology. (DOE and EPA)

Figure H-1.—Limestone Injection Multistage Burner For Simultaneous NO_x and SO_x Control

TABLE H-1 CONTROL OPTIONS FOR MAN-MADE ACID PRECIPITATION PRECURSORS

Major Acid Precipitation Pollutants	Major Sources	Applicable Control Approaches	Principles of Operation
Precursor Pollutants:			
Sulfur Dioxide (SO_2)	– Electric Utilities: primarily high-S coal & oil power plants	– low sulfur fuel switching (commercial)	– Substitute naturally occurring low-S coal or oil.
		– physical coal cleaning (PCC) (commercial)	– Remove physically bound sulfur (pyrite) from coal via separation processes prior to combustion.
	– Industrial Fuel Combustion: primarily high-S coal & oil combustion	– flue gas desulfurization (FGD) (commercial)	– React acidic flue gas with alkaline liquids yielding either a throwaway or saleable sulfur compound product.
		– limestone injection multistage burners (LIMB) (developmental)	– Inject limestone in boiler through low-NO_x burners to react with sulfur gases to produce a throwaway product.
Nitrogen Oxides (NO_x)	– Electric Utilities: coal, oil, and gas fired	– low excess air (commercial)	– Minimize air used for combustion to reduce NO_x production from air N_2.
		– staged combustion (commercial)	– Add overfire air ports to allow fuel-rich early combustion conditions to reduce NO_x to elemental N_2 and O_2.
	– Industrial Fuel Combustion: coal, oil and gas boilers	– low NO_x burners (1st gen. commercial; 2nd gen. burners in demo phase)	– Replace old burners with specially designed burners to maximize fuel-rich early flame conditions for NO_x reduction.
		– flue gas cleaning (FGC) (commercial in Japan)	– Treat flue gas with NH_3 in selective catalysts to reduce NO_x to N_2 and O_2.
	– Stationary Engines: large diesels, gas turbines, and IC engines	– catalytic reduction (demo phase)	– Utilize combustion gas catalytic systems to reduce NO_x to N_2 and O_2 either with NH_3 (fuel-lean engines) or w/o NH_3 (fuel-rich engines).
		– combustion modification (demo phase)	– Modify engine to encourage staged combustion.
	– Mobile Sources: autos and trucks	– exhaust gas recirculation (commercial since 1972)	– Recirculate exhaust gas back to cylinders to lower temp. and lower oxidation of air N_2 and NO_x.
		– three-way catalyst (commercial since 1982)	– Special tailpipe catalytic system to reduce NO_x to N_2 and O_2.
Directly Emitted Acidic Pollutants:			
Acid sulfates; predominantly sulfuric acid mist.	– Electric Utilities: coal- and oil-fired power plants	– Limited information suggests FGD and low-S fuel switching may provide some level of control for coal	– See SO_2 discussion above.
	– Non-Utility Fuel Combustion: coal and oil combustors	– Low excess air operation	– Lowering combustion air tends to decrease oxidation of SO_2 to SO_3 with subsequent sulfuric acid formation.
Hydrogen Chloride (HCl)	– Electric Utilities: high-Cl coal-fired power plants – Industrial Boilers: high-Cl coal	– flue gas desulfurization (FGD) – limestone injection multistage burners (LIMB)	– See SO_2 discussion above.

TABLE H 2 RETROFIT CONTROL TECHNOLOGIES FOR COAL FIRED UTILITY BOILER

Process	SO_2 and NO_x Removal Efficiencies	Status	Estimated* Captial Costs ($/kW)	Applicability	Problems
Flue Gas Desulfurization (lime or limestone scrubbing)	90% SO_2 Reduction	Commercial	200 to 300	Most large utility boilers; costs could sharply increase over estimated values for difficult retrofits.	– High costs, especially for difficult retrofits. – Large quantities of wet waste to be disposed of.
Physical Coal Cleaning	20 to 30% SO_2 Reduction	Commercial	40 to 50	Centralized cleaning facilities can allow broad applicability.	– Limited SO_2 removal, especially for difficult to clean coals. – Large quantities of pyrite solid residue for disposal.
Low-Sulfur Fuel Switching	60 to 80% SO_2 Reduction	Commercial	26 to 61	Most utility boilers.	– Boiler derating sometimes required. Potential displacement of locally mined coal.
Low-NO_x Burners (second generation)	60 to 70% Reduction	Early Commercialization	2 to 7	Most modern utility boilers.	– NO_x removal only; no SO_2 removal.
Limestone Injection Multistage (Low NO_x) Burners (LIMB)	50 to 60% SO_2 Reduction 50 to 60% NO_x Reduction	Developmental	27 to 37	Most utility boilers.	– Technology will be commercial no earlier than 1985. Questions must be resolved regarding performance and boiler reliability.

* Based on 300 MW, 4% sulfur coal and Dec. 1982 dollars.

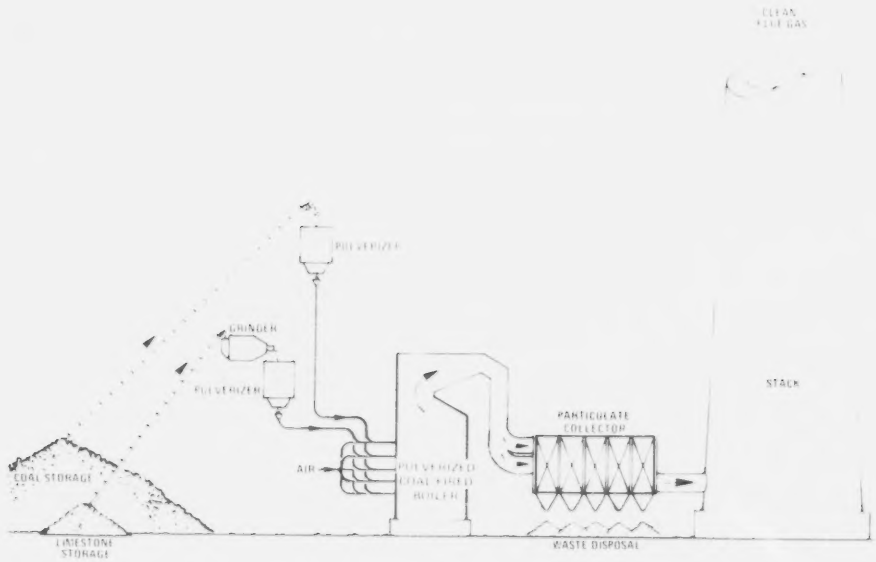
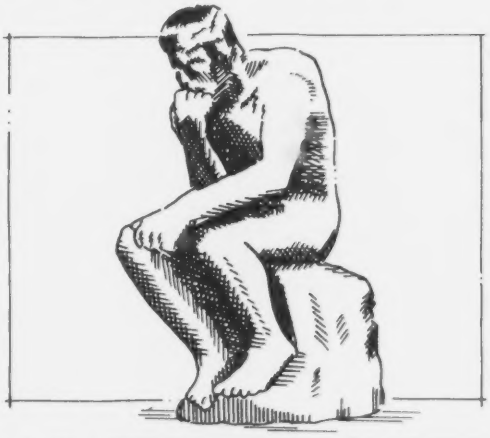


Figure H 2 Schematic of Integrated Limestone Injection Multistage Burner (LIMB) System

I. Assessments and Policy Analysis

BACKGROUND

Work on assessments and policy analysis began when the National Acid Precipitation Assessment Program started in FY 1982. While the other technical and scientific components of the National Program date back to the rise in scientific interest in acid deposition, the assessment effort is directed toward specific goals and objectives that were defined in detail only when the Task Force began planning the National Program.



Assessments and policy analysis research must produce quantitative methods that organize and display scientific information in ways that allow comparison of alternative policies. A wealth of analytic methods is currently available: decision analysis, technology assessment, benefit-cost analysis, risk analysis, and so on, but seldom have they been comprehensively applied to regional air quality problems like acid deposition.

Two primary efforts to study the implications of air quality management decisions on a regional level have recently been published. One of these, the *Ohio River Basin Energy Study*, was sponsored by the EPA under the direction of the U.S. Senate. The other, *Costs and Benefits of Sulfur Oxide Control*, was prepared by the Organization for Economic Cooperation and Development (OECD) and treated the Western European situation.

Although both studies broke new ground in significantly advancing the methods available for performing such analysis, they also recognized substantial problems as yet unresolved in producing definitive results. Chief among these are ". . . the considerable uncertainties (partly due to lack of knowledge) that exist in assessment of the different components of the sulfur problem . . . the data base is by no means well established. And, the 'science' intended to improve this situation still requires a number of years to reach maturity."(OECD, p. 8)

More recently, the Congressional Office of Technology Assessment (OTA) has undertaken a comprehensive assessment of *The Regional Implications of Transported Air Pollutants*. The July 1982 Interim Draft of this report clearly recognizes the large uncertainties in current knowledge about the causes and consequences of acid deposition. The OTA report makes a useful contribution, attempting to summarize what is and is not known and to suggest approaches to decisionmaking under uncertainty. But the report also recognizes that the type of benefit-cost analysis that is an objective of the National Program is not currently feasible in the face of these uncertainties.

RESEARCH GOALS AND ACTIVITIES

The goal of the Assessments and Policy Analysis research program is to build upon earlier studies and draw from existing analysis methods to construct the means for comprehensive benefit-cost assessments. This capability will be unique in at least two ways. First, it will be designed to deal quantitatively with the range of uncertainty around various data and their use. This will allow continuous tracking of the research program while uncertainties decrease over time. In addition, developing methods to organize scientific results and apply them to policy questions early in the research program will ensure that the National Program can produce information that is relevant to making policy decisions.

The Task Force has defined the scope of the major assessment in FY 1985 to encompass four areas: (1) an assessment of current damages attributed to acid deposition; (2) an uncertainty analysis of key scientific areas, especially emissions and atmospheric processes; (3) the implications of uncertainty in these areas on policy alternatives; and (4) a description of the framework for the integrated assessment methodology, including the status of the development of its components, that will be the basis of the 1987 and 1989 integrated assessments.

The 1985 date for the current damage assessment is based on realistic projections of how soon critical scientific information will be available. Ongoing research programs within the National Program will delineate the existing uncertainties in the science and the impact these uncertainties have on policy alternatives.

More comprehensive assessments based on an integrated framework that combines emissions models, source/receptor relationships and dose-response functions will be possible in 1987 and 1989. At present, much of the basic information for these assessments is unavailable

but research projects within the National Program are vigorously pursuing this information.

Figure I-1 shows the information flow for the current damage assessment component of the FY 1985 assessment. The deposition information, inventories of sensitive resources, and dose-response information are produced by various technical components of the research program and combined to produce an estimate of the magnitude of physical damages. The assessments research program is working toward a detailed accounting of economic damages.

Figure I-2 illustrates the framework for improving information and models of atmospheric processes and similarly improving the way analyses of ecosystems and effects come together to compare costs and benefits of alternative policies. The key parts of this framework should be available in 1986 and integrated assessment results by 1987.

The Assessments and Policy Analysis program will prepare integrated assessments of the research results produced throughout the National Program. These assessments will involve interpreting the importance and quality of the research results, developing estimates of the benefits and associated costs of alternative control and mitigation measures, and formulating guidance for policymakers.

By applying benefit-cost assessments and special studies as needed, the National Program Assessment Group expects to answer the following key questions:

1. What is the physical, biological, and economic significance of current and expected adverse or beneficial effects from the deposition of acidic and acidifying materials in North America?
2. How are the composition and distribution of acid deposition in North America linked with emission patterns, and what significance do uncertainties have for control or mitigation strategies?
3. From what existing range of strategies for integrated emission control and receptor-oriented mitigation can policymakers choose?
4. Which strategies show the greatest promise of cost effectiveness or optimal cost-benefit, and what bounds of uncertainty should be placed around such conclusions?
5. What specific research would most effectively reduce the physical, biological, and economic uncertainties decisionmakers must face in choosing among strategy options for dealing with acidic deposition?

1985 CURRENT DAMAGE ASSESSMENT

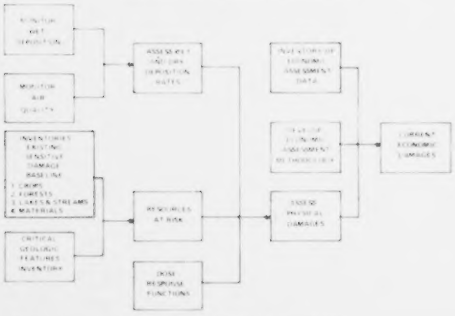


Figure I-1. Inputs for current damage assessment.

PROGRAM ACCOMPLISHMENTS—FY 1982

- **Critical Assessment Document.** This document was requested by the Clean Air Science Advisory Committee. It differs from the future assessments planned for the National Program in that it is restricted to examining the current scientific literature in terms of its scientific quality and limitations. It does not attempt a comprehen-

INTEGRATED ASSESSMENT 1987 AND BEYOND

Figure I-2. Inputs for integrated assessments.

sive interpretation of the implications of this literature. A draft of more than 1,200 pages was prepared for public scientific review workshops in November 1982. This draft is now being revised by its 55 authors and editors and will be submitted in May 1983 for printing as a public review draft.

- **Integrated Control Strategies Modeling Project.** On the basis of this research, a detailed report on sulfur dioxide emissions from utilities for 1976 through 1980 has been prepared. The data base from which the report was produced is maintained on Federal Government computers. The U.S. utility industry emissions information contained in the U.S./Canada Memorandum of Intent Working Group 3B final report has been taken from this source. This project also analyzed projected emission rates to the year 2000. These reports provide definitive information for Government policymakers on the current and projected future emission rates of relevant pollutants.

- **Development and testing of methods for uncertainty estimation.** This project, begun in 1982, will provide (1) estimates of uncertainty in models and data elements, especially important where direct validation cannot be made; (2) estimates of the overall uncertainty arising from a combination of uncertainties about a series of processes; and (3) a basis for including uncertainty in the development of control or mitigation strategy. The first phase of this effort is underway; a report in 1983 will publish the results. This report will guide policymakers on the scientific limitations to predicting the effectiveness of proposed emission control programs.

- **Sulfur and nitrogen over the eastern North American airshed.** Substantial progress was made in developing a detailed atmospheric budget for all sources of sulfur and nitrogen compounds in eastern North America. This project will produce a final report during FY 1983. An effort to develop a comprehensive mass budget for sulfur and nitrogen fluxes, through integrated atmospheric, terrestrial, and aquatic ecosystems, will build upon this work. These emission inventories are also essential data for the atmospheric transport models used to quantify source/receptor relationships to study emission control options.

- **Aquatic effects of acid precipitation.** A model was developed to estimate the dose-response relationship (between surface water, pH, and number and species of fish) for aquatic ecosystems using probabilistic damage functions. Uncertainty in this analysis was handled by interviewing experts in the specific area of aquatic biology using a structured method to include their estimates of probability or uncertainty. The model will be used to estimate the economic damages expected from the acidification of surface waters.

- **Development of integrated assessment methodologies.** Policy analysis requires a consistent method for using the many relevant data bases, models, dose-response and economic relationships needed to predict the benefits and costs of any proposed emission control or mitigation. An Advanced Integrated Assessment Methodology is being developed to link these several analytical tools and relationships. Where appropriate, uncertainties will be handled within the methodology. The development of this methodology was begun in FY 1982 and will continue over the next several years as our scientific understanding of acid deposition matures.

FUTURE ACTIVITIES

FY 1983, with the exception of the projects already discussed, is the first year for the full program in Assessments and Policy Analysis research. Assessment research and development over the next 3 years will fall into three activity blocks, as outlined in Table 1. Emphasis will be placed on the first two blocks which are aimed at the long-term development of an integrated assessment methodology and at providing mid-course corrections to the overall national research program. Additionally, preparation of the 1985 assessment will require considerable effort over this time period.

The first block of activities focuses on developing a methodology for integrated assessments. These assessments combine available information on emissions sources, atmospheric transport, and deposition of acidic and acidifying materials; their transport after deposition through the relevant physical and biological receptor systems (both terrestrial and aquatic); and their effects upon these systems with estimates of how they change in response to changes in emissions or receptor-oriented mitigation measures. An integrated assessment also includes calculated costs of such control and mitigation measures. Performing an integrated assessment on a number of possible control and mitigation measures builds up a body of data for policy analysis.

Methods for conducting an integrated assessment of the acid deposition problem require extensive development. The first block is structured to break the development work into the several major components required to meet the first three key questions listed earlier. The last element within the block assembles these components into an Advanced Integrated Assessment Methodology (a well-defined protocol by which to conduct assessments).

The second block of activities provides detailed planning, oversight, coordination, and review to construct linkages for the Advanced Integrated Assessment Methodology. These include quality assurance for the assembled data bases and validation for the required process-simulation models. Additionally, this block of activities includes the actual assembly and integration of all the analysis tools required by the Advanced Methodology.

The third block of activities applies the Advanced Integrated Assessment Methodology to developing optimal strategies for emission control and mitigation. Additionally, other intermediate assessments and short-term policy analyses are included within this block, thus addressing the last two key questions.

A framework will be developed for integrating and coordinating research and deliverables between the Assessments and Policy Analysis Task Group and the other Task Groups, including a critical path analysis of the deliverables required from each, and will define the spatial and temporal resolution required for different data sets, relationships, or models. This analysis will identify key elements of the framework which, if delayed, would slow the overall assessment process. The integrating framework will generally resemble the information flow diagrams presented in Figures I-1 and I-2.

This research framework should not be misinterpreted to imply that the activities it outlines flow sequentially from the concept of a methodology to eventual application. In actual practice, this is a dynamic process with much iteration and many feedback loops. The Task Group must develop those feedbacks early in performing the work outlined in the second and third blocks in order to guide the development work in the first block.

Major reports expected in FY 1983 include the final Critical Assessment Document, a report on sulfur and nitrogen atmospheric budgets for eastern North America, a report on uncertainty analysis methodology evaluation, and a report on probabilistic dose-response damage functions for aquatic effects of acidification.

Table 1.—Tasks Group I Research Framework

A. Integrated assessment methods development
1. Develop and apply economic benefits assessment methods;
2. Develop advanced effects assessment methodology;
3. Develop advanced methods for source/receptor assessment;
4. Develop integrated method's for constructing emission control and mitigation strategies; and
5. Develop advanced methods for integrated assessment.

B. Integrated assessment coordination and integration
1. Oversee assessment planning and coordination;
2. Coordinate data base development activities;
3. Coordinate model development and validation activities; and
4. Develop assessment model linkages.

C. Integrated assessment application and policy analysis
1. Perform and participate in special scientific/technical assessments;
2. Identify and analyze alternative emission control and mitigation measures;
3. Preliminary policy analysis/applied integrated assessment; and
4. Review status of assessments of major policy issues.

J. International Activities

BACKGROUND

The Acid Precipitation Act of 1980 requires the Interagency Task Force on Acid Precipitation to coordinate U.S. efforts with research performed in other countries. The Task Force established an International Activities Task Group, chaired by the Department of State, to encourage international cooperation on acid deposition research and monitoring.

Transboundary air pollution is of widespread international concern. The United States has actively participated in a number of international forums addressing long-range transport of air pollutants crossing international boundaries. In 1979, 34 governments, including the United States and Canada and the European Economic Community, signed the Long-Range Transboundary Air Pollution Convention under the auspices of the United Nations Economic Commission for Europe (UN/ECE).

In a separate, country-to-country agreement the United States and Canada signed a Memorandum of Intent (MOI) in 1980 to negotiate a transboundary air pollution agreement. To assist in the negotiations, joint technical Work Groups will compile all relevant data on the long-range transport of air pollution, with major emphasis on acid precipitation.

Although the MOI Work Groups and this Task Force are two different entities with distinct functions, many of the same scientists are involved in both efforts. The role of the bilateral Work Groups is to establish a mutually-agreed-upon technical basis for the U.S./Canadian negotiations on transboundary air pollution. The Task Force, on the other hand, is responsible for the long-term planning and implementation of the U.S. National Acid Precipitation Assessment Program.

RESEARCH GOALS AND ACTIVITIES

The International Activities Task Group's goals are to:

1. Encourage and facilitate productive interaction between the U.S. National Program and other nations conducting acid deposition research and monitoring activities.
2. Assist the Task Force and its Research Coordination Council in tracking international cooperative efforts.
3. Recommend to the Task Force ways to improve international cooperation and identify opportunities to work with other nations toward common research goals.
4. Inform the Task Force of activities, meetings, and developments in other nations.

The U.S.-Canada Work Groups under the 1980 Memorandum of Intent took precedence over the International Task Group's activities in 1982. The U.S.-Canada Work Groups have now essentially finished their primary tasks, and the documents are under review in both countries. Thus 1983 will see a more active role for the International Task Group, as it coordinates, tracks, and fosters cooperative research between the United States and other nations.

PROGRAM ACCOMPLISHMENTS—FY 1982

In April 1982, the Task Force and its Canadian counterpart, the Federal-Provincial Research and Monitoring Coordinating Committee met and agreed to pursue greater cooperation between the two nations' research efforts. The two research coordination groups declared that working-level contacts between scientists of both nations should be increased to develop proposals for specific cooperative research, and that such proposals would then be transmitted to the U.S. International Task Group and the Canadian RMCC Secretariat. The International Task Group and the RMCC Secretariat will serve as conduits for the two-way flow of proposals until a specific proposal is jointly approved by the Task Force and the Canadian RMCC. Following approval, cooperative research will be conducted directly by the appropriate bodies in the two countries.

FUTURE ACTIVITIES

The United States is considering several proposals for additional cooperative research with Canada. During early 1983, one or more should be ready to discuss with the Canadians as a possible joint project. It was agreed at

the Task Force/RMCC meeting in April 1982, to increase cooperative efforts on quality assurance, especially in precipitation monitoring, and these activities are being expanded.

After initially focusing on developing closer cooperation with Canada, the International Task Group will begin working to foster complementary research on acid deposition with other nations and international bodies. For example, the UN/ECE-sponsored Convention on Long-Range Transboundary Air Pollution will go into effect during 1983, and the International Task Group probably will be used to develop appropriate cooperative research projects and information exchange arrangements under that Convention.

APPENDIX I

NATIONAL ACID PRECIPITATION ASSESSMENT PROGRAM

ACTUAL EXPENDITURES
FY 1982
($ in thousands)

	EPA	DOA SE	DOA FS	NOAA	DOE	PS	DOI GS	FW	TVA	TOTAL
A. Natural Sources				600						$ 600
B. Man-made Sources	870				300					1170
C. Atmospheric Processes	3273			600	805	50			126	4854
D. Deposition Monitoring	797	253	127	700	497	75	585			3034
E. Aquatic Effects	1475		135		560	94	580	75	133	3052
F. Terrestrial Effects	1355	264	570		122	178	295		66	2850
G. Effects on Materials	250					178				428
H. Control Technologies	(10,400)*				(6200)*					(16,600)*
I. Assessments and Policy Analysis	1105				260					1365
Subtotals		517	832			575	1460	75		
TOTALS	$ 9125	1349		1900	2544		2110		325	$17,353

* Control Technologies figures not included in total. These funds for general development of SO_2 and NO_x control hardware are appropriated under other preexisting programs. The Control Technologies Task Group coordinates these efforts with the assessment and research activities of the National Program.

APPENDIX II

LIST OF TASK FORCE FY 1982 ACTIVITIES

Date	Activity	Purpose
11 25 81	RCC Meeting	Review draft material for inclusion in First Annual Report and receive guidance for final draft
12 18 81	RCC Meeting	Receive OMB FY 1983 budget passback and guidance
1 26 82	Research Coordination Council Meeting	Discuss and incorporate final changes to National Plan, review Task Group functions
2 1 82	Task Force Meeting	Review and revise final National Acid Precipitation Assessment Plan
2 23 82	RCC Meeting	Develop standard format for project descriptions in Operating Research Plan (ORP) and review draft strategic long-term framework, discuss purpose and format of Task Force's First Annual Review Meeting, discuss coordination with State and private sector efforts
4 16 82	U.S. Canada Research Coordination Meeting	Meet with the Canadian Federal/Provincial Research and Monitoring Coordination Committee to seek ways to increase bilateral scientific cooperation and to discuss mechanisms for continued coordination of efforts
4 20 82	RCC Meeting	Review and approve ORP outline, detailed planning of First Annual Review Meeting, announce mid-year FY 1982 budget report
6 2 82	RCC Meeting	Review Task Groups' draft ORP's
6 8 82	Task Force Meeting	Review OMB final comments on National Plan, discuss and revise long-term framework and ORP, approve agenda for First Annual Review Meeting, plan FY 1984 budget, examine proposed scope of work for inventory of Federal/State acid deposition projects
7 13 82	RCC Meeting	Work on integration of Task Group and NLC inputs to ORP, review status of Critical Assessment Document, agree to distribution list for National Plan
7 14 82	RCC Meeting	Define interrelationships of Task Groups' inputs and outputs, identify specific crosscutting issues to discuss at First Annual Review Meeting
8 24 82	RCC Meeting	Receive preliminary FY 1984 budget guidance from OMB, review first complete draft of ORP, finalize planning for First Annual Review Meeting, discuss inventory of Federal/State research projects, report on data management needs study
9 8 82	Task Force Meeting	Task Group leaders present initial FY 1984 budget proposals for review and guidance
9 8 10 82	First Annual Review Meeting	Assess research progress, evaluate first draft of ORP, discuss proposed research directions and priorities, examine interrelationships among Task Groups
9 21 82	RCC Meeting	Task Group leaders discuss status of FY 1984 budget, review FY 1984 budget submission to OMB
9 21 82	Task Force Meeting	Review and approve final FY 1984 budget recommendations to OMB

APPENDIX III

GLOSSARY

ACID	Acidification Chemistry Information Database
ASTRAP	Advanced Statistical Trajectory Regional Air Pollution Model
AUSM	Advanced Utility Simulation Model
CANSAP	Canadian Network for Sampling Precipitation
CAPTEX	Cross Appalachian Tracer Experiment
CEQ	Council on Environmental Quality
DOA	Department of Agriculture
DOC	Department of Commerce
DOE	Department of Energy
DOI	Department of Interior
DOS	Department of State
EPA	Environmental Protection Agency
FGD	Flue gas desulfurization
GTN	Global Trends Network
HHS	Health and Human Services
ILWAS	Integrated Lake-Watershed Acidification Study
LIMB	Limestone injection multistage burners
MAP3S	Multi-State Atmospheric Power Production Pollution Study
MOI	Memorandum of Intent
NADP	National Atmospheric Deposition Program
NAPAP	National Acid Precipitation Assessment Program
NASA	National Aeronautics and Space Administration
NBS	National Bureau of Standards
NLC	National Laboratory Consortium
NOAA	National Oceanic and Atmospheric Administration
NSF	National Science Foundation
NTN	National Trends Network
OECD	Organization for Economic Cooperation and Development
ORP	Operating Research Plan
OSCAR	Oxidation and Scavenging Characteristics of April Rains
OTA	Office of Technology Assessment (Congressional)
RCC	Research Coordination Council
RMCC	Canadian Federal/Provincial Research and Monitoring Coordinating Committee
TVA	Tennessee Valley Authority
UN/ECE	United Nations Economic Commission for Europe

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Map and text

TOTAL ALKALINITY OF SURFACE WATERS -- A NATIONAL MAP

by

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Abstract. This map illustrates the regional patterns of mean annual alkalinity of surface water in the conterminous United States. As such, it affords a qualitative graphic overview to the sensitivity of surface waters to acidification. The map is based on data from approximately 2500 streams and lakes and apparent spatial correlations between these data and macro-watershed characteristics, especially land-use.

Key Words: surface water alkalinity, sensitivity to acidification, water quality.

Introduction

The accompanying map represents the first step in a comprehensive project to identify general patterns of surface water sensitivity to acidification. The map results from the growing demand for accurate identification of acid-sensitive aquatic areas of the conterminous United States and is part of a continuing program to (1) inventory and synthesize, state-by-state, the vast quantities of relevant water quality data; (2) conduct general field surveys to fill data gaps; (3) prepare detailed regional maps and update national maps; and finally (4) conduct extensive field surveys (including biological parameters) of critically sensitive areas.

The map was developed from mean annual total alkalinity values of approximately 2,500 streams and lakes and from the apparent relationships of these data with land use and other macro-watershed characteristics such as soil type and geology. Total alkalinity is used as an index of sensitivity because it expresses the acid neutralizing capacity of water bodies and thus their relative sensitivity or tolerance to acid inputs. The ranges of our six map units were chosen to illustrate patterns of relative sensitivity on a national scale. Although there is general agreement that total alkalinity expresses acid sensitivity of surface water, there is lack of agreement on exactly where the breaking points exist between sensitive, moderately sens-

itive, and insensitive waters. Hendrey et al. (1980) considered waters not sensitive to acidification when alkalinities exceeded 500 μeq/l and of high sensitivity when alkalinities were less than 200 μeq/l. The Ontario Ministry of the Environment (1981) proposed that alkalinities between 0 and 40 μeq/l indicate extreme sensitivity and those between 40 and 200 μeq/l moderate sensitivity. Zimmerman and Harvey (1978-1979) have suggested a triad of parameters to define acid sensitivity in surface waters: pH < 6.3-6.7, conductivity < 30-40 μmho/cm, and alkalinity < 300 μeq/l.

General patterns of average sensitivities of surface waters to acidification are depicted by this map, not worst-case or best-case conditions. Our intent is to show what one might expect to find in most surface waters most of the time. Subsequent larger-scale maps of the more sensitive areas will address worst-case conditions, ranges of conditions, and significant regional and (to the extent possible) local relationships between alkalinity and geology; soils; and climatic, physiographic, and human use factors. Confidence limits for areas of greatest sensitivity will also be provided. These maps will be compiled as detailed information is gathered and analyzed.

For the present, however, there is an urgent need to understand the relative sensitivity of surface waters in different parts of the country in order to (1) provide a national perspective on the extent of the problem, (2) provide logic and/or rationale for selecting geographic areas for more detailed studies, and (3) allow more accurate regional economic assessments of acid precipitation impacts on aquatic resources.

Map Development

The data used to compile this map were selected and mapped according to several categories. Stream sites were listed separately from lakes, natural lakes were distinguished from impoundments, and both stream sites and lakes were separated into two groups -- those associated with watersheds of less than 260 square kilometers (100 square miles) and those associated with watershed areas of between 260 and 2600 square kilometers (100 and 1,000 square miles). Except in areas that were very similar in land use, physiography, and soils (e.g., the Great Plains), data associated with watersheds larger than 2600 square kilometers (1,000 square miles) were excluded. As might be expected, we found that the patterns of alkalinity values in streams were quite similar to those of lakes in the same area. As the data were being gathered and plotted, and geographical patterns of high and low alkalinities developed, collection efforts tended to concentrate on these apparent areas of greatest sensitivity.

Most of the data were obtained from the U.S. Geological Survey via STORET, an EPA computer-based water quality data storage and retrieval system. The remainder came from varied sources, principally the National Eutrophication Survey (U.S. Environmental Protection Agency, 1974, 1978a, 1978b, 1978c), the Pennsylvania Cooperative Fishery Research Unit (Arnold, 1981), and the Tennessee Valley Authority (Meinert and Miller, 1981). Although various analytical procedures were used by the various agencies [U.S. Geological Survey and the Tennessee Valley Authority, single endpoint titration to pH 4.5; National Eutrophication Survey, colorometric end point (methyl orange);

and Pennsylvania Cooperative Fishery Research Unit, double endpoint titration], the alkalinity values obtained are reasonably equivalent and, we feel, comparable for our scale of spatial analysis.

Each data point was scrutinized to insure representativeness. To accomplish this, it was necessary to keep the watershed size consistent with the relative homogeneity of major watershed features such as physiography and land use. In areas of relative heterogeneity, most of the data were associated with small watersheds (less than 260 square kilometers). Representativeness of the data was imperative for detection of spatial patterns of alkalinity, possible correlations with patterns of other characteristics, and ultimately, extrapolation of the data. To include data from sites having large watersheds of widely differing characteristics (e.g., the Willamette River at Salem, Oregon, the watershed of which includes vast contrasts in soils, geology, climate, and land use), or data downstream from major industrial or municipal waste discharges, would mask these spatial patterns.

The data were plotted on a 1:3,168,000 scale base map of the United States. Each site was represented by a small circle color-coded to approximate value. The exact value of the site was noted beside the circle, together with a designation for lake or stream. The spatial patterns of alkalinity were then compared with maps showing characteristics that are believed to be driving or integrating factors affecting alkalinity; e.g., bedrock geology and soils, land use and vegetation. Driving factors, as used in this paper, refer to those that directly affect alkalinity (e.g., geology and soils). Integrating factors, on the other hand, are considered those that reflect combinations of driving factors; for example, land use and potential natural vegetation reflect regional combinations (or an integration) of driving factors such as soils, land surface form, climate, and geology. We believe that the importance of each of these driving factors, and the hierarchy of importance relative to the combinations of factors varies from one region to another. Clarifying these regionalities is a major goal of our overall synoptic analyses; they will be addressed in the text accompanying the subsequent larger scale maps.

It became apparent early in this study that land use generally correlated with alkalinity throughout much of the United States, and particularly in the West. In general, surface water alkalinity was low in areas of ungrazed forest and high where cropland predominated. In-between types of land use generally reflected alkalinity values that corresponded to the degree of agricultural use. However, the apparent relationship between land use and alkalinity varied considerably; in some areas, particularly in the Southeast, the relationship was poorly defined or nonexistent.

Except for some localized situations, we were not able to relate geographical patterns of surface water alkalinity with geological sensitivity as depicted by bedrock or soil types. Recent studies by Kaplan et al. (1981), McFee (1980), and Hendrey et al. (1980), based on county-by-county average values, have demonstrated such correlations. Since alkalinity, in large part, is a function of the nature of the rock and soil makeup of a drainage basin (Cole, 1975), it did not appear unreasonable to expect similar results in this mapping study. The lack of correlation is probably in large part a function of study scale. Had our focus not been on the nation, but rather on a small

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region or state, possible surface water alkalinity/geology and/or soil type relationships may have been more perceptible. However, this lack of correlation may be due to one or more of several other factors. First, inconsistencies and inaccuracies in rock and soil type maps are common between, and even within, regions and between states. Second, the alkalinity in a lake or stream reflects the characteristics of both rocks and soils in the watershed. Even in small watersheds, large spatial variations in rock and soil types and depths can be found. Another confounding factor is that surface and subsurface watersheds frequently are difficult or impossible to define, particularly in areas of karst or continential glacial topography (Hughes and Omernik, 1981). Apparent surface watersheds of streams and lakes in such areas often differ greatly in area from the even more difficult to define ground watersheds.

Because of the general correlation of land use with alkalinity, the 1:3,168,000 scale base map with alkalinity values was overlayed onto a color enlargement of Anderson's Major Land Uses map (U.S. Geological Survey, 1970). When viewed on a light table, the general land use patterns and spatial relationships of surface water alkalinity to land use could be visualized. By studying these relationships and patterns, along with apparent local relationships with geologic and soil characteristics, interpretations were made and map units drawn to reflect these regional relationships.

Use of the Map

The development and usefulness of this map can best be illustrated by comparison with a more familiar graphic -- an isometric map of mean annual precipitation.[1] One should not use a precipitation map to predict the precipitation that will occur during a particular year at a given location. Rather, the map illustrates patterns of long term conditions. Few parts of the United States typically experience a truly "normal year" climatically. Generally, precipitation totals are somewhat higher or somewhat lower than the mean; occasionally, total deviation from the mean is extreme. Admittedly, precipitation maps may provide a more accurate indicator of their subject than the alkalinity map because of their more extensive data base (particularly from the temporal standpoint). However, precipitation maps are compiled using data from different geographical locations together with knowledge of apparent associations of these data with physiographic characteristics, water bodies, ocean currents, latitude, and other environmental factors. For example, precipitation patterns in mountainous areas, where data are scarce or lacking, are drawn to reflect the expected orographic effects of elevation and exposure to weather systems. Much the same kind of qualitative analysis was used to compile the alkalinity map. It is based on values from more than 2,500 stream sites and lakes throughout the United States, as well as knowledge of the apparent associations between the alkalinity data and other spatial phenomena, particularly land use.

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[1] McDowell and Omernik (1979) used this comparison to clarify the utility of a set of national maps of nutrient concentrations in streams from nonpoint sources (Omernik, 1977). The total alkalinity map was compiled in a similar fashion as the nutrient maps but with more than two and one-half times as many data points.

As with a precipitation map, caution should be exercised when using this alkalinity map. In many parts of the nation, nearly all of the surface waters have mean annual alkalinity values within the range illustrated in the map. In other areas -- particularly where there are complex variations in geology and soil type, and other factors affecting acid sensitivity -- there are wide spatial and temporal variances in alkalinity. For these types of areas, at this scale of mapping, we were only able to estimate the mean annual alkalinity of most surface waters; many may reflect higher or lower values.

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TOTAL ALKALINITY OF SURFACE WATERS*

ACID DEPOSITION
Atmospheric Processes in Eastern North America
A Review of Current Scientific Understanding

Committee on Atmospheric Transport and Chemical Transformation in Acid Precipitation

Environmental Studies Board

Commission on Physical Sciences, Mathematics, and Resources

National Research Council

NATIONAL ACADEMY PRESS
Washington, D.C. 1983

NOTICE: The project that is the subject of this report was approved by the Governing Board of the National Research Council, whose members are drawn from the councils of the National Academy of Sciences, the National Academy of Engineering, and the Institute of Medicine. The members of the committee responsible for the report were chosen for their special competences and with regard for appropriate balance.

This report has been reviewed by a group other than the authors according to procedures approved by a Report Review Committee consisting of members of the National Academy of Sciences, the National Academy of Engineering, and the Institute of Medicine.

The National Research Council was established by the National Academy of Sciences in 1916 to associate the broad community of science and technology with the Academy's purposes of furthering knowledge and of advising the federal government. The Council operates in accordance with general policies determined by the Academy under the authority of its congressional charter of 1863, which establishes the Academy as a private, nonprofit, self-governing membership corporation. The Council has become the principal operating agency of both the National Academy of Sciences and the National Academy of Engineering in the conduct of their services to the government, the public, and the scientific and engineering communities. It is administered jointly by both Academies and the Institute of Medicine. The National Academy of Engineering and the Institute of Medicine were established in 1964 and 1970, respectively, under the charter of the National Academy of Sciences.

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OFFICE OF THE CHAIRMAN

This report was undertaken by the National Academies, funded in part by a consortium of foundations*, because of the importance that we attach to illuminating the critical issues concerning acid rain. The report summarizes current scientific understanding about what is known and what is not known about relations between emissions of acid forming precursor gases and acid deposition. It should be of considerable interest to policy makers involved in regulatory decision making and in setting research priorities.

Those charged with private and public policy decisions will be particularly interested in the study Committee's evaluation of currently available scientific evidence as it affects the issue of linearity or non-linearity in relations between emissions and deposition. The public policy implications of this relationship are of major economic and political significance.

The Committee devotes a great deal of attention to discussion of areas of uncertainty that are often the result of incomplete data. But despite the uncertainties, and even though there is as yet no complete understanding of the multiplicity of complex chemical reactions and transport mechanisms, the Committee concludes that there is no evidence that the relationship between emissions and deposition in northeastern North America is substantially non-linear when averaged over a period of a year and over dimensions of the order of a million square kilometers. It is the Committee's judgment that if the emissions of sulfur dioxide from all sources in this region were reduced by the same fraction, the result would be a corresponding fractional reduction in deposition. The data reviewed by the Committee support this conclusion only in northeastern North America. The judgment is based on several pieces of evidence.

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*Carnegie Corporation of New York
Charles E. Culpeper Foundation, Inc.
The William and Flora Hewlett Foundation
The John D. and Catherine T. MacArthur Foundation
The Andrew W. Mellon Foundation
The Rockefeller Foundation
The Alfred P. Sloan Foundation

One is the well documented observation that the molar ratios of sulfates to nitrates deposited at many observation points in this region are nearly constant, and are similar to the molar ratios of the integrated emissions. The Committee believes that the simplest and most direct interpretation of this observation is the one they have given. A second observation that supports the Committee's judgment comes from a 13-year run of observations at a single location in North America that shows the linear relationship between deposition at this one site and overall emissions. Although these observations were made at a single geographic point, they represent the result of well designed and carefully taken measurements. Finally, the judgment draws upon the results of recent laboratory studies that are discussed in the report.

The Academies' process of selecting a diversified, expert study Committee and subjecting the Committee's report to a substantive review was carefully followed in the preparation of this document. In fact, because of the importance of this particular question, the review procedure took a longer time than usual and involved a larger number of critical readers than is customary.

I believe that we could not have found a more expert panel or have subjected this report to a more searching review. I hope that this report will help inform the public policy decision making process and will also permit the design of research programs that will result in more complete scientific data leading to more definitive conclusions and recommendations in the future.

Sincerely,

Frank Press

Frank Press
Chairman

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Preface

In 1981 the National Research Council issued a report titled Atmosphere-Biosphere Interactions: Toward a Better Understanding of the Consequences of Fossil Fuel Combustion. The report focused on the effects on living systems of atmospheric pollutants associated with energy production. One chapter in the report described in detail the state of knowledge concerning the effects of acid precipitation on the biosphere. Based on its survey of field data in sensitive areas and the results of experiments, the report concluded that with respect to sensitive freshwater ecosystems, "It is desirable to have precipitation with pH values no lower than 4.6 to 4.7 throughout such areas, the value at which rates of degradation are detectable by current survey methods. . . . In the most seriously affected areas (average precipitation pH of 4.1 to 4.2), this would mean a reduction of 50 percent in deposited hydrogen ions." However, a change in pH, which is measured on a logarithmic scale, from 4.1 to 4.6 corresponds to a threefold decrease in deposited hydrogen ions rather than a twofold decrease.

The conclusion was misinterpreted in the press and by others (see, for example, Science 214:38 October 2, 1981, and page A26 of The Washington Post for October 16, 1981) as a recommendation for a 50 percent reduction in emissions of the pollutant gases, sulfur dioxide and the oxides of nitrogen, that are precursors to acid precipitation. While the report concluded that the desired reduction in the deposition of hydrogen ions implies a reduction in emissions, it did not indicate how much additional control of emissions would be required to meet the goal.

As a consequence, the Chairman of the National Research Council asked the Environmental Studies Board to

review the current state of knowledge about atmospheric processes that link emissions to deposition with the purpose of describing to the extent possible the consequences of the goal of reduced deposition for emissions. By how much would emissions have to be reduced to reduce the deposition of hydrogen ions by 50 percent in sensitive areas? Conversely, by how much would depositions be reduced if there were specific reductions in emissions?

Our committee was organized in January 1982 under the auspices of the board to address this question. The work of the commitee was supported by funds provided by the National Academy of Sciences.

We conducted the study by reviewing the current literature and consulting with a number of colleagues who are knowledgeable in one aspect or another of atmospheric science related to acid precipitation. To assure an up-to-date assessment, we did not restrict our review to the literature that had already been published but included where appropriate materials that had been accepted for publication in the peer-reviewed literature although not yet actually published. Although initial drafts of various sections of our report were prepared by individuals, the document was extensively reviewed and revised by the entire committee. The report represents the collective views of the members. The appendixes were prepared by individuals to provide more detailed technical discussions than seemed warranted in the report.

We are grateful to our colleagues for their enthusiastic cooperation when we asked for their data, analyses, and views. In particular, we want to express our thanks to Perry Sampson, who modified his computer model of deposition chemistry at our request. The results are described in Chapter 3. We are also grateful to Paul Altschuller and Rick Linthurst for allowing us to incorporate material into Appendix C that originally was prepared at their request for a Critical Assessment Document on Acidic Deposition (being prepared by them under contract to the U.S. Environmental Protection Agency) and to Bruce Hicks and Jake Hales, who wrote that material. Noor Gillania and D.E. Patterson also graciously permitted John Miller to prepare Appendix B based on work they originally prepared for the Critical Assessment Document. William Stockwell carried out numerous simulations of the complex chemistry of the atmosphere, and Brian Heikes assisted with our analysis

of observational data. Ginger Caldwell helped in our review of statistical methods of analysis.

We also want to thank the members of the NRC staff who assisted in our work. Myron F. Uman, Janis Friedman, Kate Nesbet, and Janet Stoll provided staff support. Editorial assistance was provided by Robert C. Rooney, Jacqueline Boraks, Christine McShane, and Roseanne Price. The manuscript was processed under the capable and patient leadership of Estelle Miller, and the graphics were prepared by the Design and Production Department of the National Academy Press under the direction of James Gormley.

Finally, it is a pleasure to acknowledge the enthusiasm, dedication, and cooperation of my colleagues on the committee. They assumed responsibility for a difficult analysis in a highly charged political atmosphere, and they carried out that responsibility with distinction.

Jack Calvert, Chairman
Committee on Atmospheric
Transport and Chemical
Transformation in Acid
Precipitation

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ACID DEPOSITION

Atmospheric Processes in Eastern North America

A Review of Current Scientific Understanding

Summary

During the past 25 years in Europe and the past 10 years in North America, scientific evidence has accumulated suggesting that air pollution resulting from emissions of oxides of sulfur and nitrogen may have significant adverse effects on ecosystems even when the pollutants or their reaction products are deposited from the air in locations remote from the major sources of the pollution. Some constituents of air pollution are acids or become acidic when they reach the Earth's surface and interact with water, soil, or plant life. Several studies have documented the potentially harmful effects of the deposition of acids on ecosystems, which are of particular concern in areas with low geochemical capacities for neutralizing the acidic inputs (such as parts of the northeastern quadrant of North America, the Appalachian Mountains, and some of the mountainous areas of western North America). Although the pollutants may be deposited in dry form or in rain, snow, or fog, the deposition phenomenon is often called acid rain or acid precipitation. In this report we use the term acid deposition to encompass both wet and dry processes.

The question of what, if anything, to do about acid deposition is a complex one, involving generation and interpretation of scientific evidence, assessment of risks, costs, and benefits, and political considerations, both domestic and international. This report deals with a small, but important, part of the analysis that currently is being conducted to answer the question--the scientific evidence concerning the relationships between emissions of acid-forming precursor gases and deposition of potentially harmful pollutants. Our purpose is to assess the current state of scientific information that can be marshaled to describe those relationships in the

hope that our assessment will be valuable to decision makers in government and in the private sector. We focus on conditions in portions of eastern North America, for which more information is available than for other areas of the continent.

The central issues of concern in this report are the adequacy of current scientific understanding about the relationships between emissions and deposition, the extent to which the relationships are strongly nonlinear, and the extent to which distant sources contribute to deposition in ecologically sensitive, remote areas. We have reviewed the available scientific evidence that pertains to the issues of nonlinearity in the relationships between emissions and deposition and long-range transport. In the report we describe the current state of understanding about atmospheric processes (Chapter 2 and Appendixes A, B, and C), review the development of theoretical models (Chapter 3), and analyze the available observational evidence for source-receptor relationships (Chapter 4). Much remains to be learned about the detailed mechanisms involved and their relative importance for the relationships between emissions and deposition. As scientists, our training leads us to be concerned about the current limits of our understanding of the relevant processes and the uncertainties associated with assessing cause and effect. Much of our report, therefore, has been devoted to exploring the areas of uncertainty in understanding of the phenomena. Continuing research on acid deposition is needed to resolve or reduce the uncertainties and thereby to provide information useful in making more informed public-policy decisions regarding acid deposition (Chapter 5).

Our findings and conclusions are summarized below.

STATUS OF SCIENTIFIC KNOWLEDGE

Current scientific understanding of the relationships between emissions of precursor gases, such as sulfur dioxide (SO_2) and the oxides of nitrogen (NO_x), and deposition of acids or acid-forming substances, such as sulfuric acid (H_2SO_4), nitric acid (HNO_3), the anions sulfate ($SO_4^=$) and nitrate (NO_3^-), and the cation ammonium (NH_4^+), is based on theoretical considerations, the results of modeling exercises, and analysis of observational data.

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Data

Data are limited that can be used to characterize air quality, meteorological conditions, and emissions from which relationships between patterns of emissions and air quality in rural areas might be discerned in North America. Most of the historical data on air quality describe urban conditions. The most reliable information on rural conditions comes mainly from a single study in the northeastern United States, the Sulfate Regional Experiment (SURE), performed in 1977-1978. A long-term record (18 years) of reasonably reliable data on deposition chemistry is available at only one site in North America. Reliable data on regional precipitation chemistry have been collected only over the past 4 or 5 years through monitoring networks set up in the United States and Canada. There are no regional data from observations of dry deposition.

Available data on precipitation chemistry and on annual average ambient concentrations of SO_2, NO_x, sulfate, ammonium, and nitric acid indicate elevated levels of pollutants in the air and acidic substances in precipitation over much of eastern North America. Ambient concentrations are much higher than can be accounted for by emissions from natural sources on a regional scale. The geographical distributions of SO_2 and sulfate differ somewhat: SO_2 concentrations are more localized in the regions around major concentrations of sources, and ambient sulfate aerosols appear to be more widely distributed. The distributions of sulfate in precipitation are similar to those of sulfate in the ambient air. Currently the molar concentrations of nitrate and sulfate in precipitation are roughly comparable over much of the eastern United States.

Ambient concentrations of air pollutants are highly variable over time, whereas rates of emissions of the precursor gases SO_2 and NO_x are less variable. Differences in temporal behavior are due in large measure to the variability of meteorological conditions and, for secondary pollutants such as sulfate and ozone, the chemical reactivity of the atmosphere and the amount of solar radiation. Concentrations of sulfate in both the air and precipitation tend to reach their maximum values in summer in the northeastern United States; seasonal variations are less evident in the Midwest and Southeast. In many areas, ambient SO_2 and NO_x concentrations are highest in the winter, although no measurements of these

parameters in ecologically sensitive areas have been reported. Nitrate in precipitation shows much less seasonal variation in the northeastern United States.

Both ambient sulfate concentrations and sulfate in precipitation have been shown to be statistically related to aerometric parameters. For example, variations in ambient sulfate concentrations (measured at ground level) are related to variations in SO_2 and ozone concentrations, relative humidity, winds, and ventilation, whereas sulfate in precipitation is related to winds, the types and rates of precipitation, and ambient concentrations of sulfur oxides. Ambient nitrate data are not amenable to similar analyses because of uncertainties in the analytical chemical methods.

Not all sulfates and nitrates in the air or in precipitation contribute to the acidity of the air or precipitation. Acidity in solution is a function of the concentration of hydrogen ions. Some sulfate and nitrate in the air and in precipitation may be associated with cations other than hydrogen, such as ions of calcium or ammonium. Thus the acidity of deposition is the result of influences of the variety of cations and anions that may be present and in general cannot be identified with one or two anions. However, once deposited, sulfate and nitrate associated with cations other than hydrogen, such as ammonium, may still result in acidification of ecosystems as a result of biological and chemical interactions in soils and water.

Meteorological Processes

One of the greatest difficulties in establishing relationships between sources of pollution and conditions in ecologically sensitive areas is that of accounting for the influences of atmospheric processes on the behavior of pollutants. These processes include the large-scale transport of air masses, atmospheric mixing near the Earth's surface, physical and chemical reactions among pollutants and naturally present species, deposition of gases and suspended particles, and cloud processes leading to precipitation. Transport, mixing, physical and chemical reactions, and cloud processes are responsible directly or indirectly for the distribution and rate of deposition of pollutants to the ground. Our empirical and theoretical understanding of the processes is strong in some aspects and weak in others.

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Meteorological processes control the transport and dispersion of pollutants from their sources. Regions of northeastern North America that are considered to be sensitive to acid deposition are subjected to highly variable meteorological conditions, with the result that, in addition to local sources, a number of geographically widespread source regions are likely to contribute to the total deposition of acid-forming chemicals. Nonetheless, empirical analyses suggest that many of the precipitating air masses--and therefore most of the pollution-related ions dissolved in precipitation--reaching several sensitive, remote areas of the northeastern United States and southeastern Canada have their origins in upwind regions to the south and southwest. Because of the high variability in synoptic-scale meteorological phenomena affecting sensitive areas, however, all sources in eastern North America must be considered as contributing in one degree or another to the phenomenon of acid deposition. Evidence exists for long-range transport of pollutants leading to acid deposition, but the relative contributions of specific source regions to specific receptor sites currently remain unknown.

Models

Methods are available for estimating the effects of emissions of SO_2 and NO_x on regional distributions of ambient concentrations of these gases. The methods include statistical analysis of observational data and theoretical calculations using deterministic models of the chemical and physical processes involved. The methods employed to date, however, do not produce reliable estimates of spatial and temporal distributions of acidity, partly because of incomplete inventories of basic compounds in the air that neutralize some of the acidity.

The actual limitations of predictive models for calculating the effects of sources on sulfate deposition are not well defined because of deficiencies in knowledge of atmospheric processes and the lack of coherent regional data on air quality and deposition. No studies of the validity or limitations of models for nitrate concentrations have been performed. No model for wet deposition of acidity has been developed that takes account of sources and distributions of all the important ions in precipitation.

The deterministic models that are available employ by

necessity approximations to the atmospheric processes that are known or hypothesized to be important to acid deposition. All deterministic models are limited in their usefulness by the sparseness of meteorological data compared with that needed as input to project reliably the movement and mixing of air masses.

ISSUES

Applicability of Models to Decisions on Control Strategy

Results from current air-quality models applied to regional-scale processes have provided guidance on the significance of dynamic processes influencing sulfur deposition. The results of the models are qualitatively consistent with observations, thus demonstrating important temporal and spatial scales of the source-receptor relationships. Qualitatively, the models have pointed to the importance of certain geographical groupings of (SO_2) sources and the potential influence of the sources on certain receptor areas. However, current models have not provided results that enable us to have confidence in their ability to translate SO_2 emissions from specific sources or localized groupings of sources to influences on specific sensitive receptors. Little has been done in modeling to translate NO_x emissions into nitrate deposition or to link sulfate and nitrate to acid (H^+) deposition. A predictive capability that includes accounting for important cations is considered an essential requirement where long-range transport processes are involved.

Because of the simplifying assumptions made in order to develop practical, economical regional-scale air-quality models and because data are not available to validate or verify the models, workers in the field generally have only limited confidence in current results. The models and their results are useful research tools; but given the state of knowledge of the physics and chemistry of the atmosphere in the context of long-range transport in air pollution, we advise caution in using deterministic models to project changes in patterns of deposition on the basis of changes in patterns of emissions of precursor gases.

For practical purposes, deterministic models have been and will continue to be used for research on atmospheric

transport and transformation chemistry, and this use may lead to improvements in our understanding of deposition of acidifying substances in eastern North America. Before models can be relied on for development of refined strategies for dealing with acid deposition that take into account the specific locations of sources and receptors, however, we will need a greatly improved base of meteorological data and more precise treatment in the models of both chemical and meteorological aspects such as vertical distributions of meteorological variables, including clouds. In the near term, we believe that collection and analysis of field data are likely to lead to improved understanding more quickly than refinement of deterministic models. In fact, such data are needed to improve the models themselves. Confidence in control strategies will be strengthened to the extent that they are founded on scientifically sound, verified models.

Laboratory evidence suggests that an alternative model of the chemical processes involved in acid deposition may be postulated that represents the gas-phase reactions leading to the oxidation of SO_2 more correctly than the model of Rodhe, Crutzen, and Vanderpol, which has been widely used for this purpose. In keeping with results of laboratory experiments, the alternative model employs a series of reactions that results in oxidation of SO_2 without net consumption of a major oxidant, the hydroxy radical. When these gas-phase reactions are incorporated into the model, the previously reported nonlinearity in the relationship between changes in ambient concentrations of SO_2 and changes in ambient concentrations of sulfate aerosol is greatly reduced (see Chapter 3).

Nonlinearity

There is, admittedly, much to be learned about the relationships between emissions and deposition. However, on the basis of analysis of currently available data in eastern North America and within the limits of uncertainty associated with errors in the data and in estimating emissions, we conclude that there is no evidence for a strong nonlinearity in the relationships between long-term average emissions and deposition. This conclusion is based on analysis of available data on historical trends (mainly at the Hubbard Brook Experimental Forest in New Hampshire), the ratios of pollutants

in emissions and deposition, and comparison of the percentages of emissions of SO_2 and NO_x that are deposited as sulfate and nitrate in precipitation (see Chapter 4); it is also supported by theoretical calculations taking account of the latest results of realistic laboratory studies (see Chapter 3).

The only available direct evidence of strong nonlinearity between average emissions and average deposition of sulfur compounds is found in the extensive data taken in Europe over the past 25 years. Analysis of the European data has focused on historical trends in bulk sulfate deposition and on the spatial distribution of the ratio of sulfate to nitrate in monthly bulk samples. The analysis has also employed the original Rodhe-Crutzen-Vanderpol model. The observed trends in Europe are somewhat uncertain because of changes in sampling and analytical techniques and analytical laboratories throughout the period (see Chapter 4).

Reasonably reliable historical data indicating trends in North America are available only from the Hubbard Brook site (see Chapter 4). These data, from 13 years of weekly bulk samples, show no evidence of strong nonlinearity.

Analysis of the spatial distributions of the molar ratio of SO_2 to NO_x in emissions and the molar ratio of sulfate to nitrate in precipitation provides additional though indirect evidence that there is no strong nonlinearity in the relationships between long-term average emissions and deposition in eastern North America (see Chapter 4). The molar ratio of sulfate to nitrate in precipitation does not vary substantially over a large region in eastern North America; and for annual average data, it is similar to the average molar ratio of SO_2 to NO_x in emissions. Analysis of the data from the midwestern and northeastern United States also indicates that the percentage of emitted SO_2 that is deposited as sulfate in precipitation is approximately equal to the percentage of emitted NO_x deposited as wet nitrate in that region. Since the conversion of NO_2 to HNO_3 and its subsequent incorporation into cloud water are believed to be relatively rapid and efficient processes, the data suggest that the combined gaseous and aqueous conversions of SO_2 to sulfate are similarly efficient. It is therefore improbable that the oxidation of SO_2 is sufficiently hindered by a lack of oxidant to cause a disproportionately small reduction in sulfate concentra-

tions in precipitation as a result of a given reduction in SO_2 emissions.

The North American data therefore suggest that (a) whatever atmospheric processes are taking place, pollutants are being thoroughly mixed over a region with linear dimensions up to 1,000 km, and (b) the formation of sulfate is neither enhanced nor retarded relative to the formation of nitrate. The conclusion is clouded by three types of uncertainty: the limited amount and uncertain quality of the data, the natural variability of atmospheric processes, and the lack of firm understanding of the physical and chemical processes involved.

If improved measurements indicate that the relationships between emissions and deposition in Europe and eastern North America are different, the differences between the two regions in meteorology, or latitude, or other factors, such as the spatial distribution of sources, may be responsible.

Influences of Local and Distant Sources

Theoretical and observational evidence exists for the long-range transport of air pollutants leading to acid deposition (see Chapters 3 and 4). However, the relative importance for deposition at specific sites of long-range transport from distant sources as compared with more direct influences of local sources cannot be determined from currently available data (see Chapter 4) or reliably estimated using currently available models (see Chapter 3).

Trends in the historical data at the Hubbard Brook Experimental Forest appear to reflect general trends in emissions (see Chapter 4). Available meteorological analyses of trajectories of precipitating systems at three locations in the Northeast (Whiteface Mountain and Ithaca in New York and south central Ontario) indicate that much of the acidity in precipitation--as well as much of the precipitation--comes from air masses arriving from the South and Southwest.

Based on the analysis of spatial distributions of the annual average molar ratios of pollutants in emissions and deposition, it appears that the atmospheric processes in eastern North America lead to a thorough mixing of pollutants, making it difficult to distinguish between effects of distant and local sources (see Chapter 4).

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IMPLICATIONS FOR EMISSION-CONTROL STRATEGIES

The implications of our findings and conclusions for choosing among possible emission-control strategies, should they be deemed necessary, are limited. We do not believe it is practical at this time to rely upon currently available models to distinguish among alternative strategies. In the absence of other methods, analysis of observational data provides guidance for assessing the consequences of changing SO_2 emissions for wet deposition of sulfate.

If we assume that all other factors, including meteorology, remain unchanged, the annual average concentration of sulfate in precipitation at a given site should be reduced in proportion to a reduction in SO_2 and sulfate transported to that site from a source or region of sources. If ambient concentrations of NO_x, nonmethane hydrocarbons, and basic substances (such as ammonia and calcium carbonate) remain unchanged, a reduction in sulfate deposition will result in at least as great a reduction in the deposition of hydrogen ion.

It can be stated as a rule of thumb that the farther a source is from a given receptor site, the smaller its influence on that site will be per unit mass emitted. Analysis of air-mass trajectories and modeling may provide insight into the relative contributions of subregional groupings of sources to sulfate deposition in ecologically sensitive areas. Interpretation of this information, however, is subjective, and it will entail considerable judgment in assigning zones of influence of sources, even for long-term averages. This subjectivity has been a source of major differences in expert opinion, and it will continue to be until scientific knowledge improves considerably.

On the basis of analysis of the spatial distributions of the molar ratios of pollutants in emissions and deposition and assuming that all other emissions and conditions remain unchanged, we would expect that if the molar ratio in emissions in eastern North America were changed by changing SO_2 emissions, a similar change would occur in the ratio of sulfate to nitrate in wet deposition. If, as described in Chapter 4, dry deposition is linearly proportional to emissions, then the average annual ratio in total deposition in the region should also respond to changes in the emission ratio. Because the analysis is based on spatial distributions, its applicability is limited to

circumstances in which the spatial distribution of emissions is not changed. Because we cannot rely on current models or analyses of air-mass trajectories, we cannot objectively predict the consequences for deposition in ecologically sensitive areas of changing the spatial pattern of emissions in eastern North America, such as by reducing emissions in one area by a larger percentage than in other areas.

RESEARCH NEEDS

We believe that extensive laboratory, field, and modeling studies should be continued if we are to establish the physical and chemical mechanisms governing acid deposition (see Chapter 5). It appears to us, however, that useful information about the delivery of acids to ecologically sensitive areas by transport and transformation processes can be determined more quickly by direct empirical observation in the field than by other means. Although the results of such field studies may not yield complete detailed descriptions of the interactions of all the processes involved, the studies are likely to provide basic phenomenological evidence with sufficient reliability to form a basis for improving the near-term strategy for dealing with the problem of acid deposition in eastern North America. Indeed, the data are essential to enhance theoretical understanding and to develop improved deposition models. In the long term, however, the ultimate strategy for dealing with acid deposition will depend on the application of realistic, validated models.

1 Introduction

During the past 25 years in Europe and the past 10 years in North America, scientific evidence has accumulated suggesting that air pollution resulting from emissions of hydrocarbons and oxides of sulfur and nitrogen may have significant adverse effects on ecosystems even when the pollutants or their reaction products are deposited from the air in locations remote from the major sources of the pollution (National Research Council 1981, Environment '82 Committee 1982). Some constituents of air pollution are acids or become acidic when they reach the Earth's surface and interact with water, soil, or plant life. Several studies have documented the potentially harmful effects of the deposition of acids on ecosystems (NRC 1981, National Research Council of Canada 1981, Overrein et al. 1980, Drablos and Tollen 1980). Although the pollutants may be deposited in dry form or in rain, snow, or fog, the deposition phenomenon is often called acid rain or acid precipitation. In this report we use the term acid deposition to encompass both wet and dry processes.

DEPOSITION ACIDITY

An acid is a chemical substance that, in water, provides an excess of hydrogen ions (H^+) to the solution.[1] In solutions, the electrical charges of positively charged ions (cations) balance those of negatively charged ions (anions). In acid precipitation, excess hydrogen ions are usually balanced by sulfate ($SO_4^{=}$), nitrate (NO_3^-), and to a lesser extent chloride (Cl^-) ions. There may, in general, be other cations in addition to H^+ present in precipitation. Organic acids, for example, are

found in all areas, but they are important as donors of hydrogen ions only in remote areas where concentrations of sulfate and nitrate are lower (Galloway et al. 1982). Acid deposition in dry form consists of gases such as sulfur dioxide (SO_2), nitrogen oxides (NO_x), and nitric acid vapor (HNO_3) as well as particles containing sulfates, nitrates, and chlorides.

Acids occur naturally in the atmosphere because, for example, of the dissolution of carbon dioxide (CO_2) in water or the oxidation of naturally occurring compounds of sulfur and nitrogen. The "natural" acidity of rainwater, measured as pH,[2] is often assumed to be pH 5.6, which is an idealized value calculated for pure water in equilibrium with atmospheric concentrations of CO_2. However, the presence of other naturally occurring species, such as SO_2, ammonia, organic compounds, and windblown dust, can lead to "natural" values of pH between 4.9 and 6.5 (Charlson and Rodhe 1982, Galloway et al. 1982).

The presence of compounds of sulfur and nitrogen of anthropogenic origin tends to increase the acidity (lower the pH) of precipitation. More than half the acidity of precipitation averaged over the globe may be due to natural sources, but anthropogenic sources may dominate in some regions. For example, in eastern North America (i.e., east of the Mississippi River) 90 to 95 percent of precipitation acidity may be the result of human activities, although natural sources may also be important at times in specific locations (U.S./Canada Work Group #2 1982).

Figure 1.1 shows the mean value of pH in precipitation weighted by the amount of precipitation in the United States and Canada in 1980. There are no known natural causes that can account for either the distribution or the value of acidity in eastern North America. The region of highest acidity does, however, correspond to the regions of heavy industrialization and urbanization along the Ohio River Valley and the Eastern Seaboard, where anthropogenic emissions of sulfur dioxide (Figure 1.2), nitrogen oxides (Figure 1.3), and hydrocarbons are high. Figures 1.4 and 1.5 indicate the spatial distributions of sulfate and nitrate, respectively, weighted by the amount of precipitation that was deposited in North America in 1980. The data were obtained from several monitoring networks in the United States and Canada.

Trends in acid deposition in North America have been difficult to discern, and data with which to assess them are sparse. Comparisons of historical data (for example, Cogbill and Likens 1974) have been questioned because of

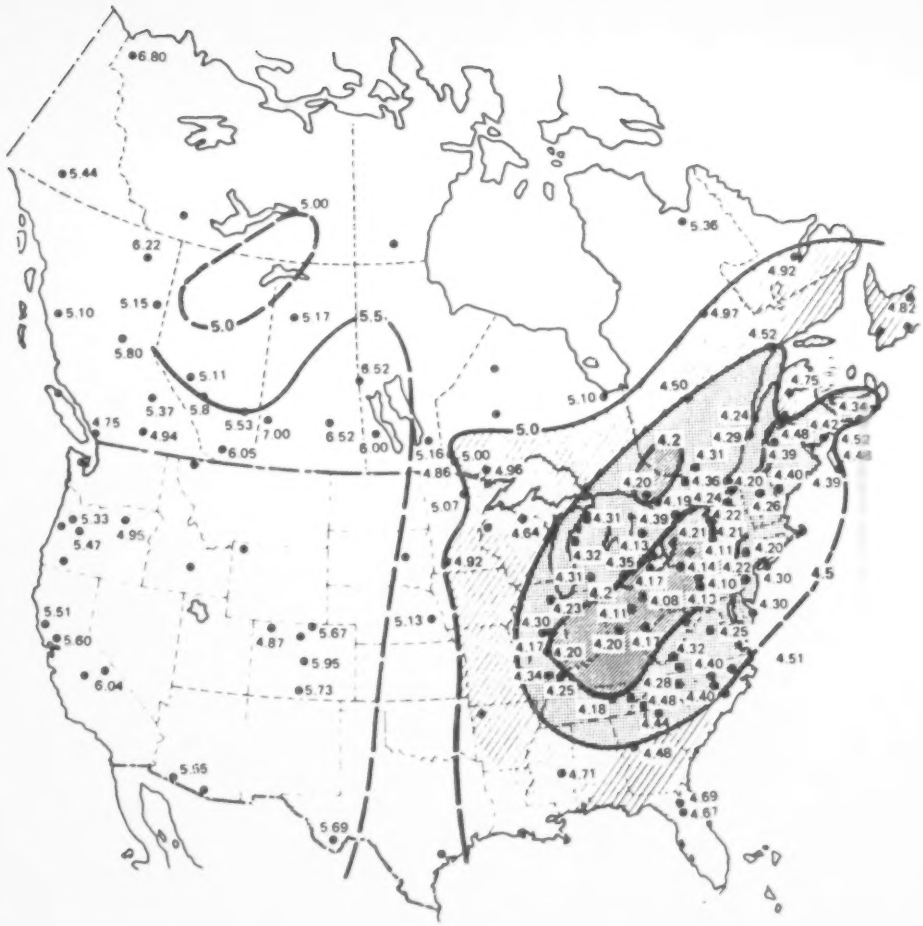

FIGURE 1.1 Annual mean value of pH in precipitation weighted by the amount of precipitation in the United States and Canada for 1980. SOURCE: U.S./Canada Work Group #2 (1982).

difficulties associated with comparing data obtained by means of different experimental methods of uncertain comparability at different sites at different times and because of difficulties in taking into account the influence of neutralizing substances on the data (Hanson and Hidy 1982, Stensland and Semonin 1982). A long-term (18-year) record of reasonably reliable data on deposition chemistry is available at only one site in North America (see Chapter 4).

The relationship between emissions and deposition in North America is complicated by changes that are not reflected in data on aggregate emissions. For example,

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because of concern about urban air pollution in the 1960s, there has been a tendency since then to build large new facilities away from urban centers and to use tall stacks to eject emissions at higher altitudes, hence promoting dispersal and dilution of the pollutants. Pollution control equipment installed during this period also changed the chemical and physical characteristics of the emissions, substantially reducing direct emissions of sulfates and neutralizing substances in fly ash. Thus, while total emissions of SO_2 in the United States increased between 1960 and 1970, urban concentrations of SO_2 decreased (Altshuller 1980). Almost all available data on air quality reflect urban conditions. Only recently have extensive networks of monitors been established in rural areas.

FIGURE 1.2 Representative values of SO_2 emissions in the United States and Canada in 1980 (thousands of metric tonnes). SOURCE: U.S./Canada Work Group #3B (1982).

FIGURE 1.3 Representative values of NO_X emmissions in the United States and Canada in 1980 (thousands of metric tonnes). SOURCE: U.S./Canada Work Group #3B (1982).

ENVIRONMENTAL EFFECTS

Atmospheric deposition involves three components: emissions, deposition, and effects on receptors. Certain aspects of the effects of atmospheric deposition are of particular importance for the development of effective policies for emission control. They concern the significance for receptors of (1) physical and chemical states of deposited materials and (2) rates and reversibility of acidification.

In discussing these issues, it is helpful to distinguish between primary, secondary, and tertiary receptors according to their proximity to the initially deposited material. Primary receptors experience direct contact with atmospheric pollutants. Examples are the surfaces of

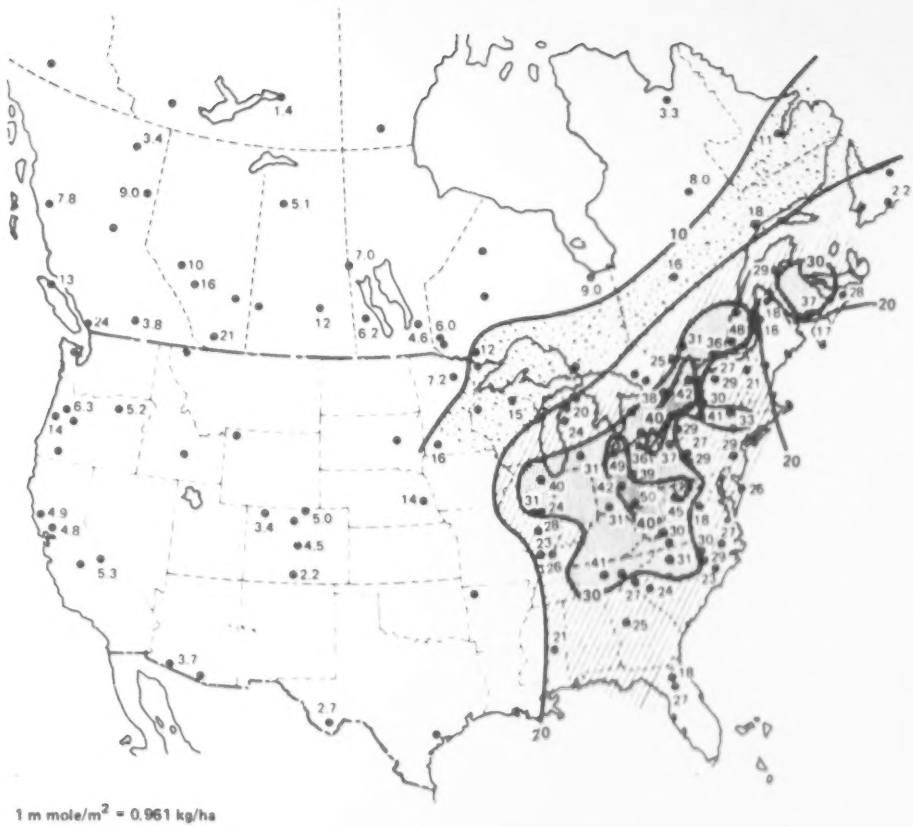

FIGURE 1.4 Spatial distribution of mean annual wet deposition of sulfate weighted by the amount of precipitation in North America in 1980 (mmoles/m^2). SOURCE: U.S./Canada Work Group #2 (1982).

structures and materials, the outer foliage of vegetative canopies, and the surfaces of soils that are not protected by vegetative canopies. Secondary receptors are subject to wet and dry deposition indirectly and only after the pollutants have been in contact with other materials. Examples include the inner foliage of vegetative canopies, soil underneath vegetation, and subsurface layers of exposed soils. Tertiary receptors are even further removed from the point of initial contact with deposition. Examples are subsoil, underlying rock formations, streams and lakes that receive most of their water from runoff from the watershed, and lake and stream sediments. The rates of transfer and mixing of materials are affected by the proximity of the receptors to the point of initial deposition and by their mass and other physical and chemical properties.

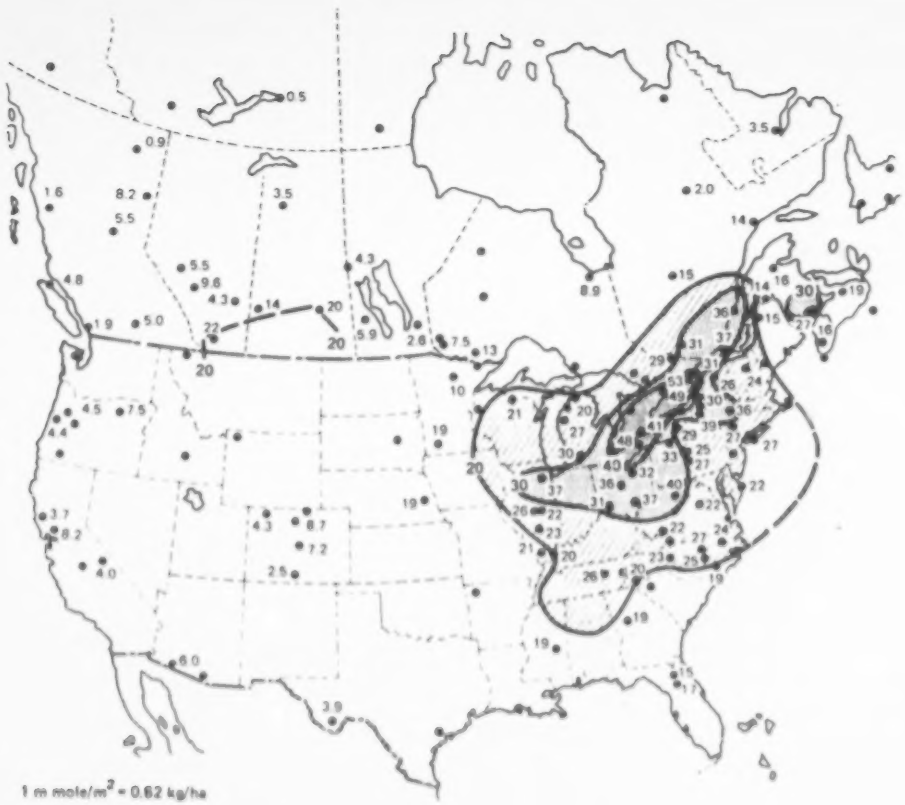

FIGURE 1.5 Spatial distribution of mean annual wet deposition of nitrate weighted by the amount of precipitation in North America in 1980 (mmoles/m^2). SOURCE: U.S./Canada Work Group #2 (1982).

Physical and Chemical States of Deposited Materials

The effects of atmospheric deposition on primary receptors depend on the physical state (solid, liquid, or gaseous) and the chemical state (e.g., sulfur or nitrogen species) of the deposited materials (NRC 1981). The physical form of deposited material determines its availability for reaction, whereas its chemical form determines its reactivity. Tertiary receptors are less responsive to the physical and chemical form of atmospheric deposition than primary and secondary receptors because of dilution. The effects of acid and acidifying ions (hydrogen, sulfate, nitrate, and ammonium) are dependent in part on the accompanying rates of deposition of neutralizing cations

(calcium). Hydrogen ions are harmful to the extent that the receptors cannot prevent or compensate for changes in acidity or the consequences of acidification.

Many chemical compounds of both sulfur and nitrogen are naturally present in soils and are involved in chemical and biological transformations in soils and vegetation. Sulfur and nitrogen are essential nutrients required for plant growth. There are many differences in the properties and biological action of the compounds of the two elements, and there are differences in the types of transformations they undergo in the environment. Biological processes (e.g., metabolic action, decomposition) have a great influence on nitrogen transformations, while both geological processes (e.g., weathering) and microbial transformations strongly affect the sulfur cycle. There is a larger pool of endogenous nitrogen than of sulfur in organisms, and larger amounts of nitrogen than sulfur are required for plant growth. The two nutrients are closely related, so that addition of one element to an ecosystem allows greater biological utilization of the other (Turner and Lambert 1980). The optimum molar ratio of sulfur to nitrogen in terrestrial ecosystems is approximately 0.03.

Nitrogen usually is efficiently metabolized in undisturbed ecosystems (Likens et al. 1977), while sulfur frequently is not retained by forest soils (Abrahamsen 1980). For aquatic ecosystems, therefore, sulfur is more important for acidification than nitrogen. Alkaline as well as acidic cations accompany the movement of sulfate from soils to aquatic systems; consequently, acidification of soil is more likely to occur from excessive sulfate deposition than from excessive nitrate deposition. The spring flush of acids into aquatic systems may, however, be closely associated with the accumulation of nitrate and sulfate in snowpack (Galloway and Dillon 1982, McLean 1981).

One of the important effects of acidification is the potential mobilization of elements from soils due to increased solubility and subsequent uptake by vegetation or movement to aquatic systems. Aluminum is present in bound form in many soils, and it can be dissolved and become available for accumulation by organisms to which it can be toxic. Dissolution of aluminum or other metals depends on the amount of water passing over a surface; solubility generally is enhanced in an acidic solution. Thus heavy rainfall exceeding surface evaporation--even with low acid content--can mobilize ions over time. This mobilization from the soil may be enhanced when acid-forming materials also are deposited from the atmosphere and washed away.

However, insufficient understanding of the interactions between soils and groundwater currently precludes estimation of the rate of mobilization of metals.

Major factors determining effects of atmospheric pollutants on forests are (1) the chemical nature and loading of deposited elements, (2) the ion exchange characteristics of soils, (3) the residence times and hydrological pathways of water through the watershed, (4) the nature and extent of existing vegetation, and (5) the geochemical activity of bedrock and soils (Evans et al. 1981, Zinke 1980). All forest ecosystems are not expected to respond to acid deposition in the same way. Effects are likely to be site-specific and dependent on the relative contributions of external and internal sources of acidity.

Major factors determining effects of atmospheric pollutants on lakes and streams are (1) the total loadings of particular compounds, (2) hydrological pathways through the terrestrial systems upstream of the water body, (3) the ion-exchange characteristics of soils in terrestrial systems upstream, (4) the residence times of water in the terrestrial systems, and (5) the geochemical reactivity of the bedrock and soils of the terrestrial systems.

Reversibility and Irreversibility

Ecosystems are repeatedly stressed by natural disasters, extreme climatic and meteorological events, and human influences such as changes in land use and pollution. Responses may be reversible or irreversible, depending on the stress, the receptor, and the time span of interest. For example, a river may carve a new channel after a flood, an effect that may be considered irreversible except by human intervention. Over a period of hundreds of years, however, the channel may fill with silt, so even this effect can be "reversed." A lake may become turbid with sediment and organic matter after a heavy rain, an effect that usually is reversed rather rapidly by natural processes. So the consequences of extreme events often are reversed by natural processes over time; as a result, considerations of the reversibility or irreversibility of effects of acid deposition should take account of the time span of interest.

The most common effect of stress on an ecosystem, such as may be caused by exposure to pollutants, is retrogression to conditions typical of an earlier stage of ecological succession. Reduction in species diversity and simplifica-

tion of ecosystem structure are typical responses to pollutants (Whittaker 1975). These changes often are accompanied by alterations in productivity. When exposures to pollutants are reduced or eliminated, natural processes may return systems either to their previous pathways of succession or to different successional pathways.

The extent to which effects of acid deposition are reversible depends on the receptor and the type of effect. Although there have been no clear demonstrations of effects on terrestrial systems to date, it is reasonable to believe that adverse effects on primary receptors are more readily reversed than those on tertiary receptors. For example, the yield of one annual crop might be reduced by the contact of acidic deposition with foliage or flowers, but a subsequent crop may be less severely affected if less acid is deposited. Damage to trees and perennial plants, particularly those that retain foliage for several years (most conifers), may be less easily reversed because of the long period required for regeneration and recovery of most woody plants. When both the aquatic and the terrestrial ecosystems are acidified in an area in which rates of mineralization and decomposition of organic matter are low, reversibility is unlikely.

Acid deposition results in net accumulations of certain elements and net losses of others in ecosystems over long time scales; the identity of the elements in each category and their rates of change vary with the ecosystem and rates of deposition. The effects of slow but persistent changes may not be apparent for many generations. Signs of these changes may be observed, but the time scales for occurrence of irreversible changes are difficult to predict, because the processes that produce and consume hydrogen ions and the reactions that affect the accumulation and loss of elements are complex and poorly understood. Extensive regions of North America (Figure 1.6) and northern Europe have little geochemical acid-neutralizing capacity. Perhaps only in retrospect will we know with certainty that systems have changed, and the reversibility of these effects by natural processes might require far more time than the period initially required to cause the changes due to anthropogenic acidification.

OTHER RELATED REGIONAL AIR POLLUTION PHENOMENA

In addition to the atmospheric processes affecting acid deposition, there are other regional air pollution

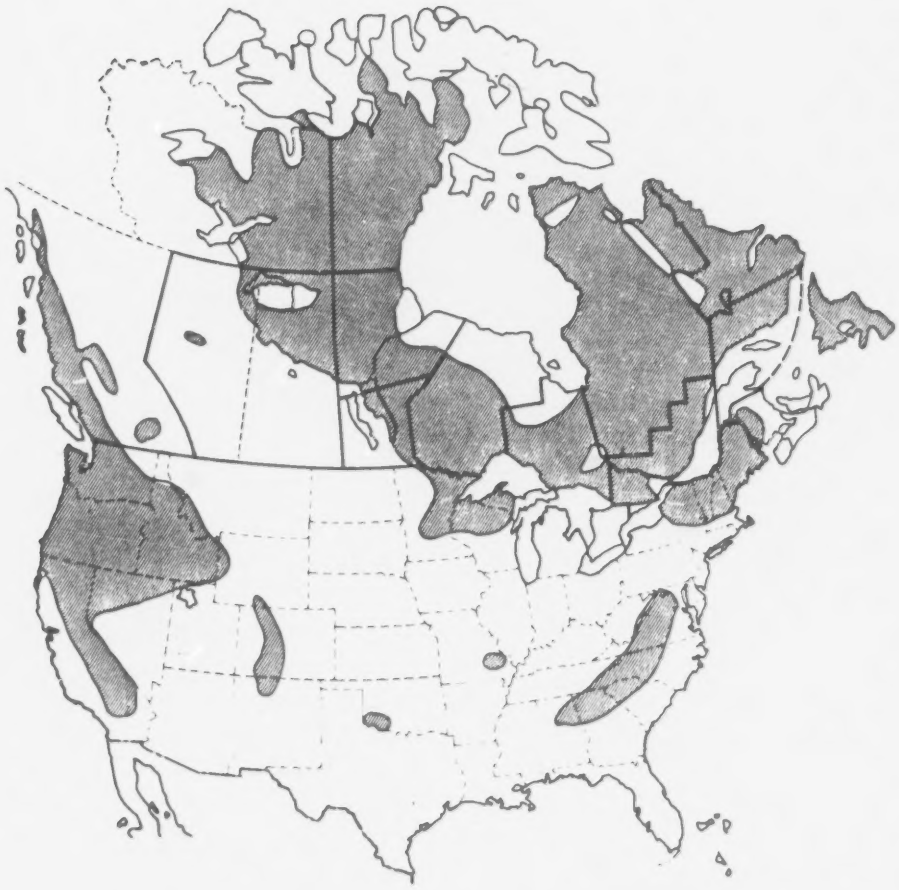

FIGURE 1.6 Regions of North America with low geochemical capacity for neutralizing acid deposition. SOURCE: Galloway and Cowling (1978).

phenomena of consequence for environmental quality that are related to acid deposition in that they are the result of similar chemical and physical processes acting on the same pollutants.

One is the occurrence of elevated concentrations of ozone (O_3) in polluted air masses extending over several hundred to a thousand kilometers (Vukovich et al. 1977, Wolff et al. 1977). Episodes of elevated ozone occur in summer under conditions that also lead to high atmospheric concentrations of sulfate aerosol, which is eventually removed by precipitation. The events are believed to be associated with increased concentrations of precursor gases, such as nitrogen oxides and hydrocarbons, that undergo reactions to form oxidants under conditions of large-scale atmospheric stagnation.

A second related phenomenon is the impairment of visibility by optically dense haze extending over large geographic areas (Trijonis and Shapland 1979). The phenomenon, which has been recognized in the eastern United States for some time, occurs during episodes of high relative humidity. Degradation of visibility has also been observed in the West (Macias et al. 1981, Trijonis 1979) and in the Arctic (Rahn and McCaffrey 1980). Although the optical characteristics of the atmosphere are linked to natural climatic factors, such as relative humidity, Husar and Patterson (1980) found an apparent association in the historical record of changes in visibility with changes in the combustion of fossil fuels. There seems little doubt that sulfate aerosols and other fine particles play a significant role in regional haze (NRC 1980). Haze in the Arctic in winter has been attributed to long-range transport of air masses polluted with sulfates and particulate carbon from sources in northern Europe (Rahn and McCaffrey 1980).

PURPOSE OF THE STUDY

The question of what, if anything, to do about acid deposition is a complex one, involving generation and interpretation of scientific evidence, assessment of risks, costs, and benefits, and both domestic and international political considerations. This report deals with a small but important part of the analysis that is currently being conducted to answer the question--the scientific evidence concerning the relationships between emissions of precursor gases and deposition of potentially harmful pollutants. Our purpose is to assess the current state of scientific information that can be marshaled to describe those relationships in the hope that our assessment will be useful to decisionmakers in government and in the private sector.

The impetus for our work has been proposals for the adoption of policies to control emissions of sulfur dioxide and nitrogen oxides (beyond current limitations on emissions from new facilities) as a means of reducing acid deposition and hence alleviating reported and anticipated damage from that deposition. The operators of sources of the pollution (mostly electric utilities, industrial boilers, and motor vehicles) reasonably wish to ensure that the costs they--and their customers--would bear as a result of control policies are commensurate with any benefits to

be obtained. A critical link in the evaluation of benefits is the estimation of the reduction in the deposition of acids that would accompany a reduction in emissions. A recent report by the National Research Council (NRC 1981) concluded that current rates of deposition of hydrogen ions should be reduced by about 50 percent (corresponding to an increase in pH of 0.3 unit) if sensitive regions of eastern North America are to be protected from adverse effects of the deposition. If this goal were adopted, by how much would emissions have to be reduced? Conversely, by how much would deposition rates be reduced if there were specific reductions in emission rates?

Our committee was organized to assess current scientific understanding about atmospheric processes that might be applied to answering these questions. Our objective was to determine what conclusions can be drawn from the state of knowledge late in 1982 about the relationships between emissions and deposition. Essential facts we faced in our work are that the subject under study is complex, the scientific evidence is evolving, and uncertainties in current understanding remain. Nevertheless, our goal required that we take account of both the theoretical understanding and observational evidence that are available today and make our best scientific judgment about their meaning.

If national policy on acid deposition is to be made on the basis of the scientific information currently available, that policy could take several forms, including maintaining the status quo. Other policies might incorporate uniform reductions (rollback) in emissions, might be designed to achieve the maximum possible environmental benefit, or might be carefully engineered to bring risks, costs, and benefits into optimal balance. The different options require scientific and technical information in different degrees of detail. In addition, they all, to one degree or another, must account for uncertainties in understanding. Decisions on almost all issues of public policy--including military affairs, the economy, and social welfare no less than environmental issues--are routinely made in light of uncertainties in knowledge. Provided uncertainties are taken into account, sufficient information is available for deciding what, if anything, to do about acid deposition.

Recognizing that uncertainties in scientific understanding about acid deposition currently exist and that uncertainties are likely to exist to some degree into the future, we believe that, whatever the near-term decision on

acid-deposition policy, research and development should proceed with the goal of supporting more advanced and sophisticated future policies. Laboratory and field research on atmospheric processes will be extremely important in this effort. In the meantime, it seems prudent to adopt policies that are flexible and adaptable to the changing base of scientific understanding. As research continues, it can be hoped that our ability to design carefully constructed optimal strategies will continually improve.

ORGANIZATION OF THE REPORT

This report describes the state of knowledge as of the end of 1982 regarding the atmospheric processes relating emissions of precursor gases and acid deposition. It does not include a detailed examination of the effects of acidic or acidifying substances on ecosystems once deposited. For such a treatment, see NRC (1981).

Chapter 2 is a general review of the current theoretical understanding of the major atmospheric processes involved in acid deposition: transport and dispersion, chemical transformation, and deposition. More complete reviews are contained in the appendixes. Chapter 3 is a general review of the theoretical models currently used or proposed for assessing the relationships between sources and receptors. Chapter 4 reviews and analyzes field data on deposition in order to develop a phenomenological understanding of acid deposition in North America. Needed research is described in Chapter 5. The focus of the report is on conditions in portions of eastern North America, for which more information is available than for regions elsewhere on the continent.

NOTES

1. More precisely, acidity in aqueous solutions is a function of the concentration of the hydrated hydrogen ion (H_3O^+), which is also called the hydronium ion. For convenience, we adopt the conventional notation, referring to H_3O^+ as H^+. In solutions, the product of the molar concentration of H^+ with that of the hydroxide ion (OH^-) is approximately constant (about 10^{-14} at 25°C). As acid is added to water, the concentration of H^+ increases and that of OH^- decreases so that the product remains

constant. By an excess concentration of H^+, we mean that the concentration of H^+ is greater than that of OH^-.

2. The acidity or alkalinity of a solution is measured on the pH scale. A solution that is neutral (neither acidic or alkaline) has pH 7.0. Decreasing pH indicates increasing acidity. The pH scale is logarithmic (pH equals the negative logarithm to the base 10 of the hydrogen ion concentration), so a solution of pH 4.0 is 10 times more acidic than one of pH 5.0.

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2 Atmospheric Processes

Physical and chemical processes in the atmosphere determine the fates of emissions of precursor gases and hence the exposures of primary receptors to pollutants. In this chapter we review current understanding of these atmospheric processes in light of the need to characterize relationships between emissions and deposition. For convenience, we consider these atmospheric processes as occurring in a sequence of clearly defined steps (Figure 2.1). The separate processes are as follows:

I. Transport and mixing
II. Chemical reactions in the homogeneous gas phase (dry reaction)
III. Dry deposition
IV. Attachment
V. Chemical reactions in the homogeneous aqueous phase (wet reaction)
VI. Wet deposition

Heterogeneous chemical processes may occur between gases and liquids adsorbed on solid surfaces, although these are generally considered to be less important in the development of acid deposition than the homogeneous processes. We therefore do not consider heterogeneous processes in this report.

Each of the separate processes takes a certain amount of time; the sum of the processing times along any particular pathway is the source-receptor transport time for the pollutant along that pathway. The processing times are extremely variable, depending strongly on meteorological processes, ambient conditions, and the presence and concentrations of various chemical species. Many of the steps are reversible, so that itinerant

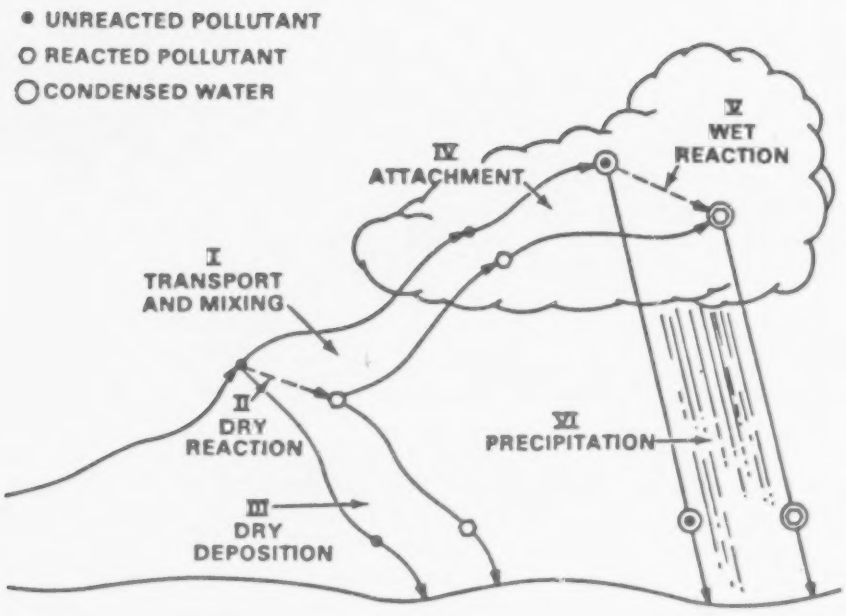

FIGURE 2.1 Atmospheric pathways leading to acid deposition.

pollutant molecules may undergo repeated cycling with corresponding lengthening of the effective processing times.

TRANSPORT AND MIXING

The transport of pollution by normal atmospheric advection and mixing is a vitally important influence on deposition phenomena, and it works both directly and indirectly. Transport phenomena directly determine where the pollution goes before it is deposited and therefore affect the atmospheric residence time of pollutant materials. Dry deposition, for example, is often limited by the speed at which the atmosphere can vertically transport pollution to the proximity of the surface.

Transport can indirectly affect pollutant deposition in a number of ways. Transport processes, for example, bring pollution into contact with storm systems, where precipitation scavenging occurs. Transport also can introduce pollutants into environments more (or less) conducive to transformation chemistry. This complex of interactions links transport with the other processes shown in Figure 2.1.

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It has been the usual practice to divide atmospheric transport processes into two categories. The first, usually termed advection, pertains to the net motion of a parcel of air as it drifts with the mean wind. The second category, diffusion, pertains to the intermixing of the parcel with its surroundings. Historically the distinction between atmospheric advection and diffusion has not been totally clear. Quite often, for example, atmospheric transport models incorporate diffusionlike terms to account for time-averaging of meandering plumes, when in fact the physical processes described have little to do with actual intermixing of materials. Similar treatments often arise in transport models using grids to approximate the desired solutions numerically. Advection processes occurring on scales smaller than the grid spacing escape resolution by the system and thus are often lumped in terms of pseudo-diffusion processes (see Appendix B).

Such approximations are often inescapable. They do, however, contribute significantly to the uncertainty in our ability to model atmospheric pollution, and they obscure the meaning of diffusion in such processes. It is therefore important to remember that advection and mixing are indeed distinct transport phenomena that can lead to different behavior of parcels of polluted air.

The distances associated with pollution transport obviously depend strongly on how long the pollutant resides in the atmosphere and thus is available for action by the advection-diffusion process. In this context it is important to note that atmospheric residence times for typical power plant pollutants (sulfur compounds, for example) are rather uniformly distributed; some pollutant molecules are deposited from the atmosphere relatively quickly and thus at locations near the source, whereas others are deposited more slowly and thus much farther away. On the basis of the best current estimates, it is not unusual for the transport distance of a given pollutant molecule to be of the order of hundreds or even a thousand kilometers. It also is not unusual, however, for a molecule to be deposited close to the source. From this one can conclude that while long-range transport processes certainly are important, shorter-range phenomena are occurring as well.

Another factor that must be taken into account in assessing transport is the height at which pollutants are released into the atmosphere. One approach to local air-quality problems has been to increase the height of

stacks in accordance with the notion that the higher the point of release the less the pollutants would affect the surrounding area. This approach to pollution control was applied to large new plants where taller stacks were constructed. At older plants, relatively short stacks were replaced with taller ones. A number of factors, such as meteorology and terrain, influence how the height of an individual stack affects dispersal of a given pollutant, so it is difficult to evaluate the effectiveness of tall stacks for dispersal in general.

Recently Koerber (1982) studied a set of 62 coal-fired power plants in the Ohio River Valley. He developed a measure of the potential for long-range transport that involved physical stack height, plume rise, and mixing height. Figure 2.2 shows the temporal trend in Koerber's parameter between 1950 and 1980. The implication of this and other work is that stack heights must be taken into account when assessing source-receptor relationships involving long-range transport.

Because of cumulative uncertainties, the trajectories and times associated with long-range transport are much more difficult to estimate than their shorter-range counterparts. Early, very crude attempts to simulate long-range phenomena simply employed local wind roses and straight trajectories from the sources in question. The obvious deficiencies associated with this approach prompted further efforts to develop curved-trajectory simulations, which were driven by conceptualized, time-evolving wind fields.

The curved-trajectory approaches, while representing a major advancement over their straight-line predecessors, suffered from two major disadvantages. The first of these was that the data from which the wind fields were derived were usually extremely sparse in both time and space--a problem that becomes particularly severe under complex meteorological conditions involving fronts and storm systems. Although a variety of sophisticated interpolation techniques has been advanced subsequently to offset this problem, the poor coverage of meteorological data in both space and time remains particularly troublesome.

The second major problem associated with these types of trajectory approaches is caused by mass motions of air vertically and vertical wind shear, i.e., the dependence of wind speed and direction on altitude. Early trajectory simulations, based on constant-altitude wind fields, soon were replaced by layer-averaged or constant-pressure

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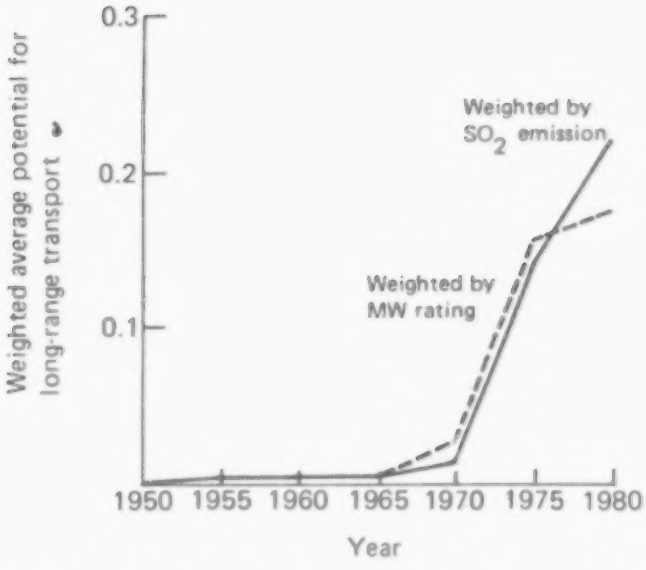


FIGURE 2.2 Trend in long-range transport potential for 62 sources in the Ohio River Valley. SOURCE: Koerber (1982).

surface versions to overcome this disadvantage partially. On the basis of thermodynamic arguments, it is expected that vertical motions of air parcels should adhere rather closely to constant-entropy surfaces in the atmosphere, and from this a few "isentropic" trajectory simulations have evolved as well. However, vertical motions caused by the heat released or absorbed during cloud formation are not taken into account by either method.

Although curved-trajectory simulations can produce rather reliable results in simple meteorological situations, they are fraught with uncertainty when conditions become complex, such as near frontal systems. Some idea of this uncertainty may be gained from Figure 2.3, which shows the results of two different calculations of a trajectory under the same conditions in the vicinity of a frontal storm. One calculation (solid curves) uses the assumption of isentropic transport, while the other (dashed curves) employs isobaric transport. After 24 hours, the calculated positions of the two hypothetical air parcels are several hundred kilometers apart (also see Chapter 3 on the treatment of transport and mixing in models as well as Appendix B).

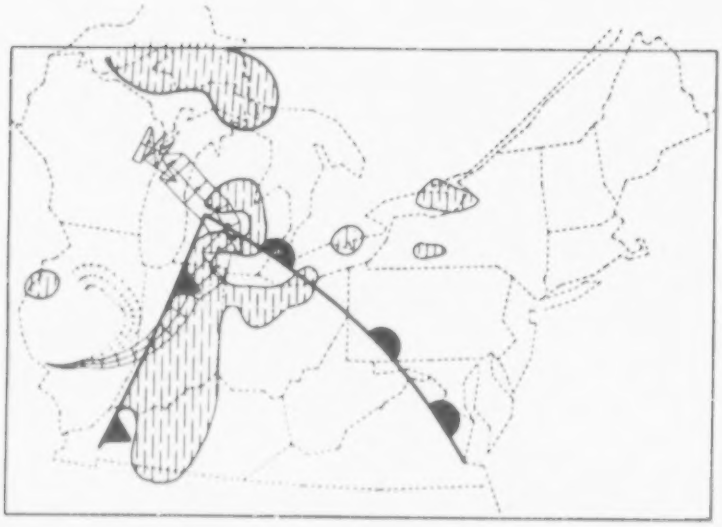


FIGURE 2.3 Calculated plume trajectories in the vicinity of frontal systems for 24 hours after release. Solid plumes were calculated on the assumption of isentropic vertical motion; dashed plumes were calculated on the assumption of isobaric motion. Shading indicates area of precipitation. SOURCE: Adapted from Davis and Wendell (1977).

The difficulties associated with wind shears and vertical motions could be largely overcome if the vertical motions were indeed known. Several weather prediction models produce such information, albeit prognostic in nature, rather coarse in scale, and dedicated more to upper regions of the troposphere than to the planetary boundary layer. In addition, a few mesoscale dynamic models currently exist that can supply initial estimates, at least, of information of this type. The application of such techniques for pollution trajectory simulation has been comparatively limited owing to the complexity of the modeling process and the expense of the simulations. A summary of several of the trajectory simulation techniques discussed here appears in Chapter 3 (Table 3.2). They have been applied to form composite regional pollution models.

A major factor contributing to the uncertainty in long-range trajectory simulations stems directly from our current inability to measure long-range transport. Several balloon studies have been attempted, but they have been less than satisfactory because of technical difficulties, the balloons' supposed inability to track

vertical motions exactly, and statistical problems associated with tagging a stochastic system with too few units. Chemical tracers have not been particularly successful to date owing to detection difficulties over large distance scales. Tracer techniques are evolving rapidly, however, and it is not unreasonable to expect some highly significant results to emerge from experiments with tracers in the next 5 years. For example, during the summer of 1983 six releases of tracers are to be made from Ohio and Ontario under the Cross Appalachian Tracer Experiment (CAPTEX). A wide arc of measurement sites will be set up over 600 km downwind of the releases. This experiment will be the first step in a long-range tracer program.

An important feature of the long-range transport of air pollutants is that the plumes from individual sources may become so dilute and so thoroughly mixed far downwind of major source areas that the attribution of specific parcels of polluted air to specific sources is impractical. In these cases, the sources contribute pollutants to air masses that may be considered to be entrained in synoptic-scale meteorological systems. The classic example of mixing occurs in large stagnant air masses that occur most frequently in summer in the eastern United States (see Chapter 4). The motion of air masses on the synoptic scale may be important for understanding acid deposition in areas remote from major source regions. The average flows across North America are shown in Figure 2.4, which illustrates that the region in which acid deposition is currently thought to be an environmental problem is also a region of intense interaction between tropical marine and arctic air masses.

CHEMICAL TRANSFORMATION

During transport through the atmosphere, SO_2, NO_x, hydrocarbons, and their oxidation products participate in complex chemical reactions that transform the primary pollutants into sulfates and nitrates. The transformation processes are important because, as we discuss later, deposition of the primary pollutants and that of their transformation products are governed by different processes.

There are many chemical pathways through which SO_2 and NO_x in the atmosphere can be transformed (oxidized) into sulfate and nitrate compounds, including homogeneous

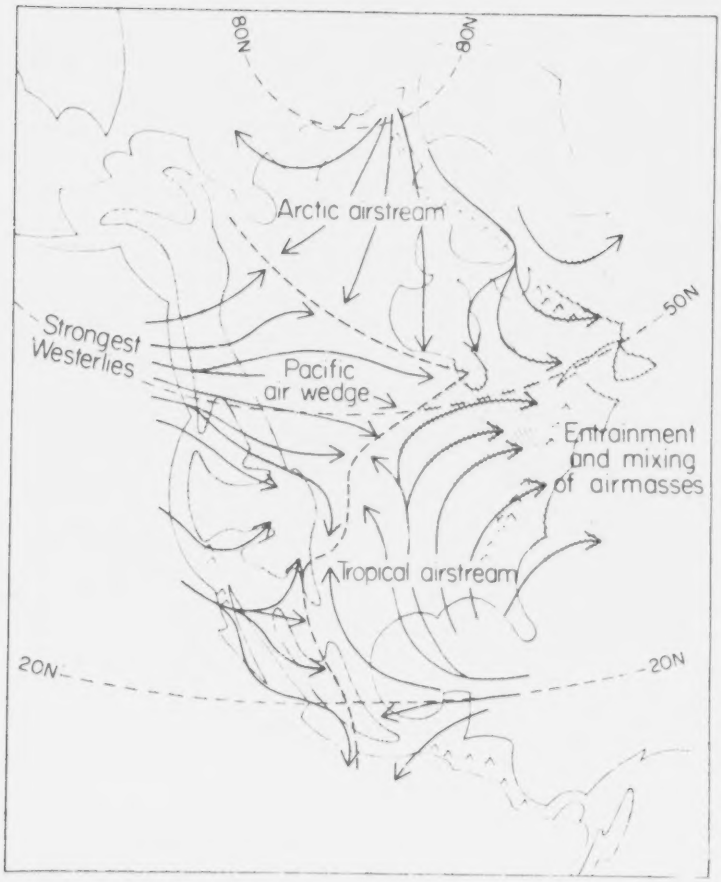
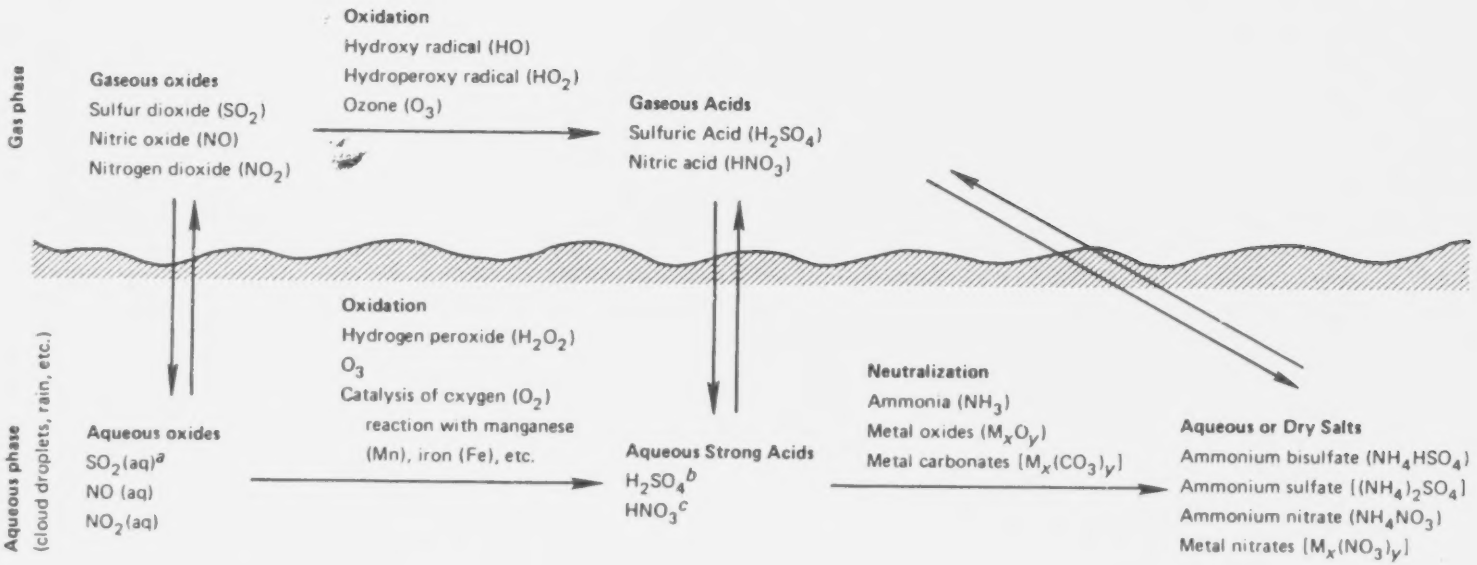


FIGURE 2 4 Surface flows across North America, illustrating the area of complex entrainment and mixing of air masses in the eastern portion of the continent. SOURCE: Bryson and Hare (1974).

processes that take place in the gas phase and in liquid droplets or heterogeneous processes that take place on the surfaces of particles or droplets. Figure 2.5 indicates the pathways by which SO_2 and NO_x are transformed into gaseous- and aqueous-phase acids. Field studies indicate that the relative importance of gas- and liquid-phase reactions depends on meteorological conditions, such as the presence of clouds, relative humidity, intensity of solar radiation, and the presence and concentrations of other pollutants.

A comprehensive review of homogeneous gas- and solution-phase atmospheric chemistry associated with acid deposition is presented in Appendix A. The appendix



[a] $SO_2(aq)$ reacts to form hydrogen ions (H^+) and bisulfite ions (HSO_3^-).
[b] Aqueous H_2SO_4 dissociates into hydrogen ions, bisulfate ions (HSO_4^-), and sulfate ions ($SO_4^=$).
[c] Aqueous HNO_3 dissociates into H^+ and nitrate ions (NO_3^-).

FIGURE 2.5 Pathways for the formation of atmospheric sulfate and nitrate.

includes detailed descriptions of alternative oxidation pathways and analyses of reaction rates. General descriptions and conclusions, drawn from this material, are presented below.

Homogeneous Gas-Phase Reactions

SO_2 and NO_x have been observed in the atmosphere to be oxidized through homogeneous gas-phase reactions at rates of a few and 20 to 30 percent/h, respectively (Step II in Figure 2.1). The observed rates cannot be explained by direct oxidation by atmospheric oxygen, reactions that occur too slowly for typical concentrations of pollutants and, in the case of SO_2, in the absence of catalysts. Similarly, although there are direct pathways to the formation of sulfuric and nitric acids beginning with absorption of solar radiation by SO_2 and NO_2, respectively, these processes also appear to be unimportant under typical conditions in the troposphere.

According to current understanding, most of the gas-phase chemistry in the lower atmosphere that results in oxidation of SO_2, nitric oxide (NO), and nitrogen dioxide (NO_2) entails reactions with a variety of highly reactive intermediates--excited molecules, atoms, and free radicals (neutral fragments of stable molecules)--that are generated in reactions initiated by the absorption of solar radiation by trace gases. The most important of the intermediates for gas phase oxidation appears to be the hydroxy radical, HO.

The hydroxy radical can be formed in the troposphere by a number of reactions. A common process begins with dissociation of NO_2 by absorption of sunlight, which forms a highly reactive oxygen atom that combines quickly with a diatomic oxygen molecule to form the triatomic oxygen molecule, ozone (O_3). Ozone may be photodissociated, yielding an electronically excited diatomic molecule of oxygen and an electronically excited oxygen atom, $O(^1D)$, which reacts readily with a water molecule to form HO. The hydroxy radical, unlike many radicals that are fragments of complex molecules containing carbon, does not react readily with molecular oxygen; HO survives in the atmosphere to react with most impurity gases, such as hydrocarbons, aldehydes, NO, NO_2, SO_2, and carbon monoxide (CO). Reactions between HO and several impurity gases produce additional classes of reactive transient species, which, in turn, react with

atmospheric constituents to form additional reactive species. For example, reactions of HO with CO and hydrocarbons produce peroxy radicals; peroxy radicals react rapidly with NO to form NO_2 and alkoxy, acyloxy, and other HO radicals.

The net result of all of these interactions is a large number of chemical pathways for oxidation of SO_2 and NO_x to sulfuric acid (H_2SO_4) and nitric acid (HNO_3), respectively, many of which depend initially on the formation of HO. A sequence of these reactions can be constructed in which a single HO radical may oxidize CO, hydrocarbon, or aldehyde, followed by oxidation of NO to NO_2 accompanied by production of additional HO radicals. Repeated cycling of the sequence results in continued oxidation of NO to NO_2 and relatively constant concentrations of HO.

There are a number of gaseous-phase chemical reactions between SO_2 and reactive transient species that may lead to formation of H_2SO_4; these reactions, along with currently accepted values for the reaction rates, are listed in Appendix A. While many of the rate constants are known only with an uncertainty of 50 percent, it appears as if the most important reaction is that between SO_2 and HO, yielding $HOSO_2$. Evidence is good that this reaction ultimately leads to the generation of sulfuric acid, and a number of pathways for this subsequent reaction have been explored. Which of these pathways is most important is still unknown, but it is likely that the oxidation of SO_2 by HO is a chain-propagating reaction.

The principal agents for oxidizing NO to NO_2 are ozone and peroxy radicals, whereas NO_2 is oxidized to HNO_3 by a well-characterized reaction with HO (Appendix A).

According to current understanding, then, the rates at which sulfuric and nitric acids are formed in homogeneous gas-phase reactions depend on ambient concentrations of the hydroxy radical. Direct measurement of HO in the atmosphere is difficult, but both theoretical and experimental estimates are available from which to estimate rates of conversion from SO_2 and NO_2 to H_2SO_4 and HNO_3, respectively. Using the rate constants listed in Appendix A, we find that for high concentrations of HO--characteristic of polluted summer sunny skies--SO_2 will be converted to H_2SO_4 by reaction with HO at a daily averaged rate of about 0.7 percent/h (16.4 percent per 24-h period), whereas NO_x

is converted at an average rate of about 6.2 percent/h (100 percent per 16-h period). At HO concentrations typical of winter sunny weather in a polluted atmosphere, the average rates are roughly 0.12 and 1.1 percent/h (3 and 25 percent per 24-h period), respectively. These rates, of course, depend on HO concentration and therefore fall rapidly after sunset since HO is formed largely by photochemical processes.

While contributions to oxidation of SO_2 from other reactions may be important in some circumstances, the rates reported in Appendix A are consistent with those observed in urban plumes in the absence of clouds. Observed conversion rates for NO_2 in cloud-free conditions are also consistent with the estimates presented in Appendix A.

Homogeneous Aqueous-Phase Reactions

Oxidation of SO_2 is rapid in water, often of the order of 100 percent/h. Rates of aqueous-phase oxidation of SO_2 are typically much higher than those of gas-phase oxidation. The lifetimes of individual clouds, however, are short, so that the long-term average oxidation rate in cloudy air may be similar to that in the gas phase. When SO_2 dissolves in water, several species are formed: the hydrate $SO_2 \cdot H_2O$ and the ions HSO_3^- (bisulfite), $SO_3^=$ (sulfite), and H^+ (hydrogen ion). As the concentration of H^+ increases, a solution becomes more acidic, corresponding to lower values of pH. The concentration of total dissolved sulfur, designated S(IV), in water in equilibrium with gaseous SO_2 at a specified partial pressure is inversely related to the concentration of H^+ in the solution. That is, SO_2 is less soluble in more acidic (lower pH) solutions. As indicated in Appendix A, equilibrium between gas-phase SO_2 and total dissolved sulfur is established quickly, so S(IV) in cloud droplets or liquid aerosol particles is a function of pH and the ambient gaseous concentration of SO_2.

Current evidence suggests that two agents, hydrogen peroxide (H_2O_2) and ozone (O_3), may be primarily responsible for oxidizing S(IV) to H_2SO_4 in atmospheric water for typical concentrations of pollutants (step V in Figure 2.1). Other possible oxidation pathways exist, including homogeneous reactions involving the hydroxy radical or aqueous-phase NO_3 and catalytic reactions involving soot or ions of manganese and iron. Current

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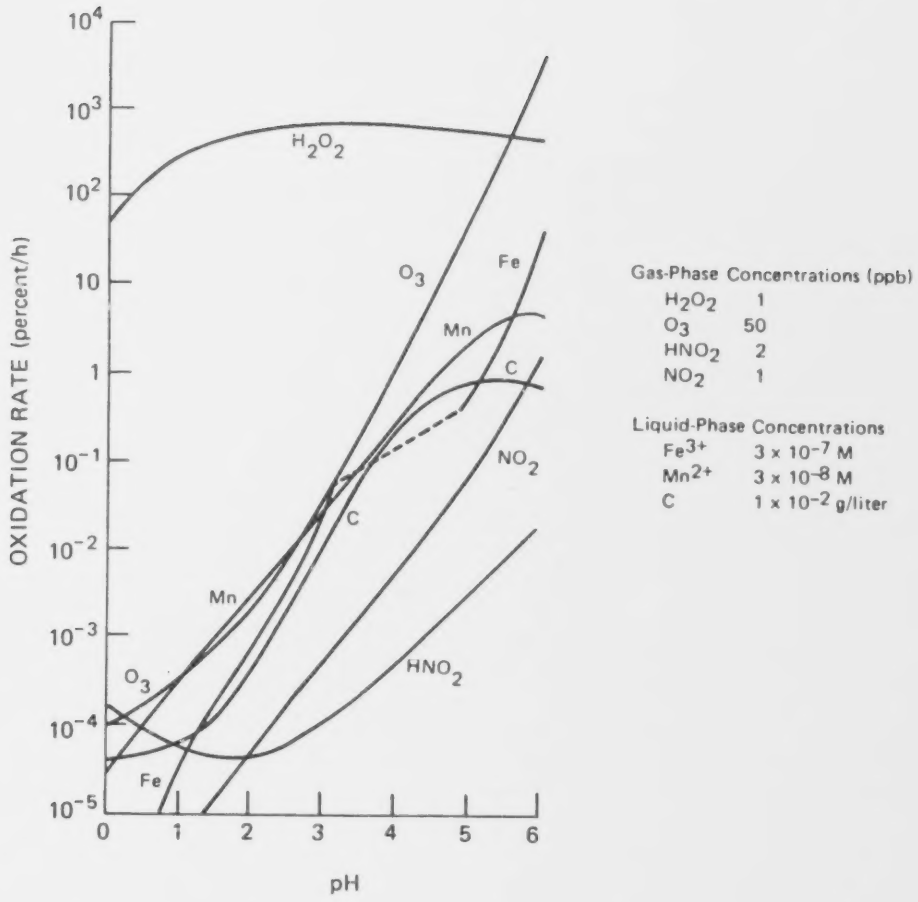

FIGURE 2.6 Theoretical rates of liquid-phase oxidation of SO_2 assuming 5 ppb of SO_2, 1 ml/m^3 of water in air, and concentrations of impurities as shown. SOURCE: Martin (1983).

theoretical understanding of the oxidation rates of dissolved SO_2 by the various proposed mechanisms for typical impurity concentrations is shown in Figure A.13 of Appendix A and reproduced here as Figure 2.6. The figure shows that oxidation by H_2O_2 predominates under the specified condition except for high values of pH (low acidity), in which case O_3 may be an important reactant as well. Because the pH of aerosol droplets and cloud water is generally measured to be below 5, H_2O_2 is currently regarded as the most important oxidizing agent in the aqueous-phase chemistry of the formation of sulfuric acid.

As demonstrated in Figure 2.6, oxidation of S(IV) by H_2O_2 is an effective process that is relatively independent of pH. The other mechanisms that have been studied are strong functions of pH; they are likely to be active early in the process of acidification of droplets but rapidly become ineffective as acidification proceeds. The relative positions on plots such as Figure 2.6 of the curves for the less important mechanisms depend somewhat on the assumed concentrations of the impurities.

Both H_2O_2 and to some degree O_3 have their origins in homogeneous gas-phase reactions. The chemistry of ozone production was described earlier. The major homogeneous sources of H_2O_2 in the troposphere are reactions involving the hydroperoxy radical (HO_2). In the polluted atmosphere there is strong competition for reaction with HO_2 among NO, NO_2, aldehydes, and other species, so the efficiency with which H_2O_2 is generated is a complex function of the concentrations of these and other impurities (see Appendix A). In theory, the amount of H_2O_2 formed in gas-phase reactions and taken up in cloud water and precipitation is sufficient to oxidize a large fraction of S(IV). Ozone is also readily taken up by atmospheric water, although its solubility is considerably lower than that of H_2O_2.

If homogeneous air masses containing H_2O_2, O_3, and SO_2 encounter aqueous aerosol droplets, cloud water, or precipitation, solution-phase oxidation of the SO_2 is favored because of the high conversion rates of these reactions. However, the optimal conditions for formation of H_2O_2 and O_3 in the troposphere differ, so the relative importance of the two aqueous-phase oxidants will depend on conditions in the gas phase that determine the relative rates of production of the two oxidants. For example, as detailed in Appendix A, conditions of low NO_x concentrations and high levels of hydrocarbons and aldehydes favor formation of H_2O_2, whereas high concentrations of NO_2, hydrocarbons, and aldehydes favor O_3 production.

While H_2O_2 is thought to play an important role in oxidizing aqueous phase SO_2 in the atmosphere (Figure 2.6), it has only recently become possible to measure concentrations of H_2O_2 reliably in ambient air or in cloud water or precipitation, because of deficiencies in experimental techniques.

In comparison with current understanding of various pathways for the formation of H_2SO_4 in atmospheric water, little is known about the solution-phase chemistry

that results in formation of nitric acid in aerosols, cloud water, or precipitation. Both theoretical and experimental evidence, described in Appendix A, suggest that dinitrogen pentoxide (N_2O_5) formed in gas-phase reactions between ozone and NO_2 may be efficiently scavenged by water droplets to form nitric acid directly. Sufficient data are not yet available on which to base evaluations of the importance of this or other mechanisms to the formation of HNO_3 observed in the atmosphere.

Most clouds evaporate before precipitation can develop. Therefore cloud processes can affect the chemical nature of sulfur and nitrogen compounds in the absence of precipitation and can contribute to their redistribution in the troposphere.

Relative Roles of Gaseous- and Aqueous-Phase Chemistry

In recent years the role of aqueous-phase chemistry in the development of acid deposition has received increased attention. The results of field and laboratory studies suggest that although rates of oxidation of SO_2 in the gas phase are relatively slow, the relative importance of gas-phase and solution-phase oxidation varies, depending on a variety of meteorological conditions, such as the extent of cloud cover, relative humidity, presence and concentrations of various pollutants, intensity of solar radiation, and amount of precipitation. Although solution-phase conversion rates can be considerably higher than those in the gas phase, air masses over the eastern United States are likely to be relatively free of clouds and precipitation a large fraction of the time, so both gaseous- and aqueous-phase processes must in general be regarded as contributing to acid formation. It is also clear from the discussion in Appendix A that formation of sulfuric and nitric acids in liquid aerosols, cloud droplets, and precipitation depends on gas-phase reactions to supply the necessary reactants.

The clearest evidence that gas-phase reactants contribute to solution-phase formation of acids was obtained in the Acid Precipitation Experiment (APEX) described briefly in Appendix A. In this experiment, the constituents of dry air were measured prior to the time the air mass ascended to produce a large area of precipitation characteristic of a warm front. Measurements were also made of samples of cloud water and of precipitation at the base of the cloud. The results indicated that

both nitric and sulfuric acids were formed rapidly in the cloud, although the oxidizing agent remained unidentified because of weaknesses in the analytical methods.

Data from another experiment are now available showing an appreciable rate of conversion of SO_2 to H_2SO_4 at night in clouds over coastal waters, indicating an oxidation process other than reaction with the hydroxy radical, which is present in significant concentrations only in daytime.

The importance in atmospheric chemistry of aqueous-phase processes taking place in clouds is illustrated theoretically in Figure 2.7, which gives the results of calculations that combine homogeneous gas-phase chemistry with the current picture of aqueous reactions (Environmental Research & Technology, Inc., and MEP, Inc. 1982). The figure shows the progress of oxidation in clear air (beginning with NO and NO_2 concentrations of 10 ppb, concentration of reactive hydrocarbon vapors of 200 ppb, SO_2 concentration of 5 ppb, and $SO_4^=$ concentration of 2 $\mu g/m^3$) and the effects of introducing a cloud with 1 g/m^3 of liquid water at 1400 h. In theory the insertion of cloud water causes dramatic decreases in atmospheric concentrations of H_2O_2, HNO_3, SO_2, and $SO_4^=$. The behavior of NO, NO_2, O_3 and peroxyacetylnitrate (PAN) was not strongly influenced by the presence of cloud water. The example demonstrates that clouds have the potential to dominate chemical interactions involving water-soluble or water-scavengable constituents. Field experiments are required to determine if this dramatic effect actually occurs in the atmosphere.

DEPOSITION

Dry Deposition

The term dry deposition is used to denote a variety of processes by which pollutant gases and aerosol particles reach the Earth's surface, including the surfaces of both living and inanimate objects on the ground (vegetation and buildings, for example). The processes depend on concentrations of the pollutants and small-scale meteorological effects near the surface as well as on the characteristics of the receiving surface.

Superficially, dry deposition seems to be almost trivially simple in comparison with other aspects of the relationships between emissions and deposition. Dry

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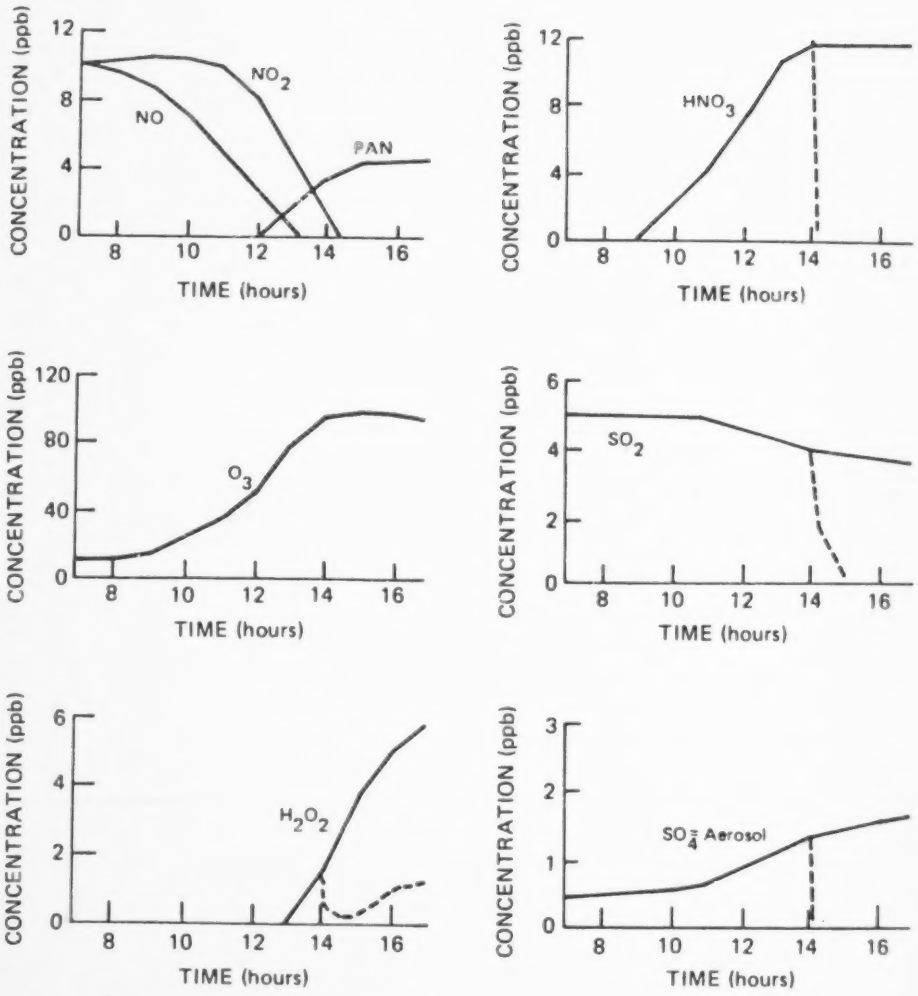

FIGURE 2.7 Theoretical calculations of gas and aerosol concentrations as a function of time for gas-phase reactions only (solid line) and with the introduction of cloud water (dashed line) at 1400 hours. SOURCE: Environmental Research & Technology, Inc., and MEP, Inc. (1982).

deposition takes place at the Earth's surface and thus is inactive in the volume of the atmosphere in which chemical transformation and processes leading to wet deposition occur.

In fact, however, dry deposition is incompletely understood. Uncertainties in dry deposition may be an important source of error in today's regional modeling efforts.

There are several reasons for the current uncertainties in understanding dry deposition (Appendix C). The first is that dry-deposition rates are extremely difficult to measure. Although a number of possible techniques exist (Hicks et al. 1981) and considerable effort has been devoted to developing appropriate methods for measuring fluxes to surfaces, the base of high-quality data is still distressingly small. Furthermore, the more reliable data that do exist tend to have been obtained under experimentally convenient conditions (for example, high pollutant concentrations, uniform terrain) and thus reflect only a small subset of the potentially important environmental conditions.

A second reason for uncertainty in dry-deposition rates is a consequence of the complexity of the physical processes in the atmosphere. As indicated in Figure 2.8, several mechanisms convey pollutants to the surface, and it is often not clear which processes dominate under which conditions. Especially important in this regard are the near-surface mechanisms for aerosol particles, such as inertial impact, phoresis, and electrical effects. Uncertainties in this area are currently substantial, especially for deposition to surfaces of vegetation.

The third reason arises from uncertainties in the characteristics of the substrate on which materials are deposited. Contrary to the superficial view that dry deposition is purely a surface phenomenon, phenomena both at and in the substrate can play a role in determining the deposition flux. It is well known, for example, that stomatal openings on leaf surfaces influence the deposition of gases such as SO_2 and ozone. Soils and building materials have been shown to "saturate" with depositing gases. Re-emission of sulfur compounds from plant surfaces has been detected. All of these results render the concept of a simple boundary condition approach to dry deposition somewhat questionable; the corresponding uncertainties are again large.

These difficulties combine to give a number of widely varying estimates for the temporal and spatial scales of dry deposition of specific pollutants. As a rule of thumb, for sulfur and nitrogen compounds at least, dry deposition is taken on the average to be about as effective as wet deposition in pollutant removal. About one third of sulfur emissions is transported out of the continent. Thus roughly one third of northeastern emissions is assumed to be dry-deposited on the North American continent (see the section in Chapter 3 on material balance).

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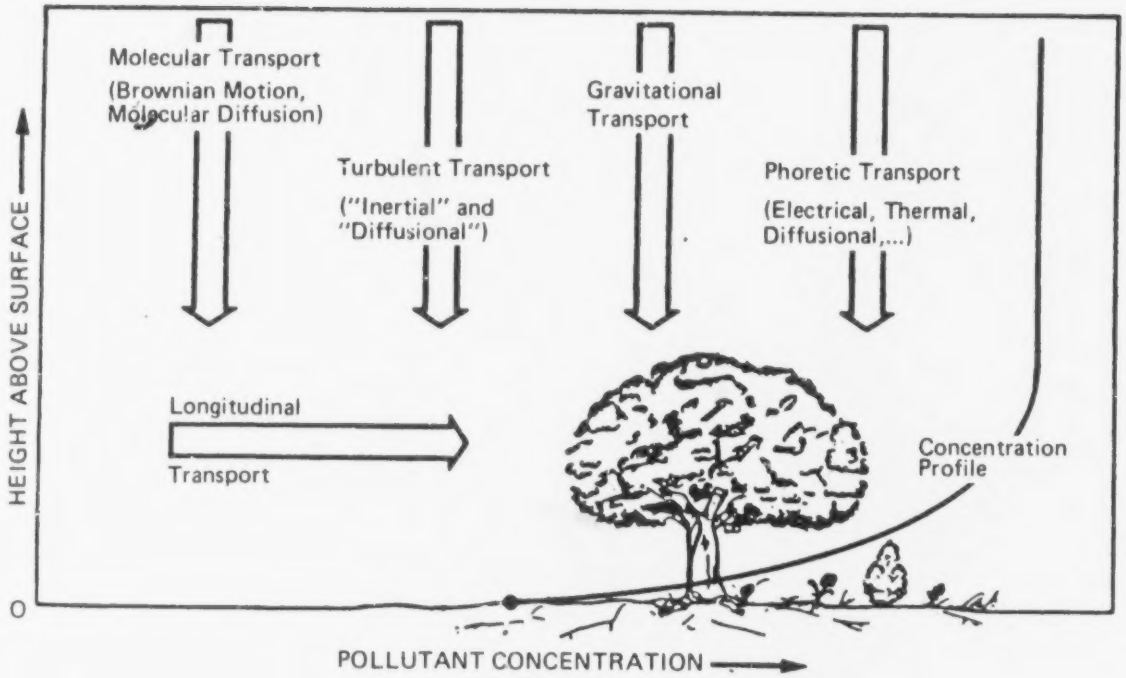

FIGURE 2.8 Pathways for dry deposition.

There are also studies, however, that obtain a scale length for dry deposition in excess of 10^4 km for some species (Slinn 1983), strongly suggesting interaction with global circulation patterns. This work is in concordance with observations of deposition in Greenland and the Arctic, as well as the general haze buildup in the northern hemisphere. Until the extent of such long-range transport is more thoroughly understood, the modeling of dry deposition is likely to remain highly uncertain.

Wet Deposition

The term wet deposition encompasses all processes by which atmospheric pollutants are transported to the Earth's surface in one of the many forms of precipitation: rain, snow, or fog, for example. Wet deposition therefore involves attachment of pollutants to atmospheric water and includes chemical reactions in the aqueous phase as well as the precipitation process itself. Aqueous phase chemical processes (step V in Figure 2.1) have been discussed previously; here we address only the physical processes by which pollutants first become attached to water droplets and then are deposited in wet form (also see Appendix C).

A rough indication of the significance of wet deposition on a continental scale can be obtained from a map of annual precipitation in the United States (Figure 2.9). From the distribution, one would expect that wet deposition would be an important contribution to total deposition in the East and in the Pacific Northwest. In regions with frequent precipitation, wet deposition also becomes relatively more important than dry deposition far away from sources, where SO_2 is depleted and sulfate particles are a significant fraction of the atmospheric sulfur burden. This also appears to be the case in remote areas of southeastern Canada.

Attachment Processes

The physical processes by which pollutants become attached to droplets and other falling hydrometeors such as ice crystals (step IV in Figure 2.1) have been the subject of extensive research, and a number of technical

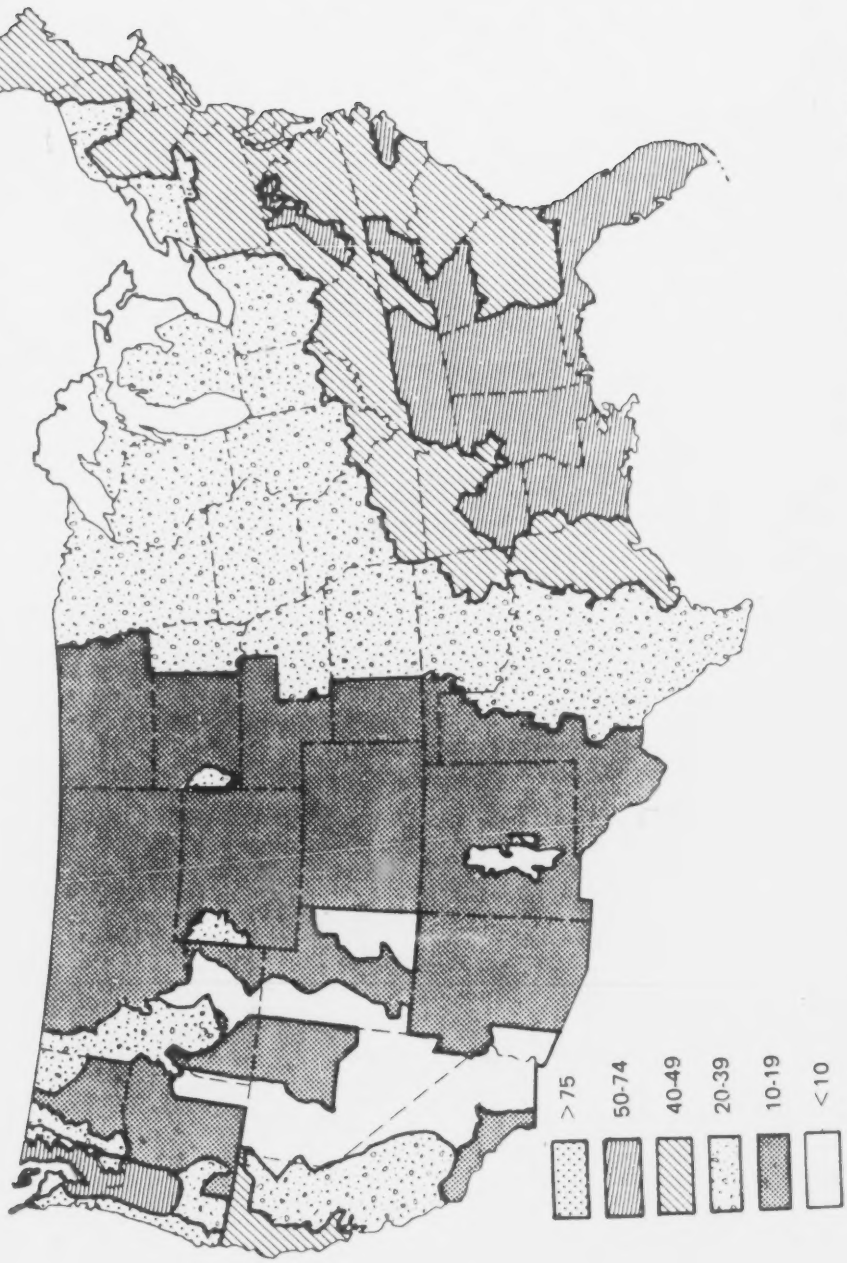

FIGURE 2.9 Average annual precipitation in the United States. SOURCE: GCA Corporation (1981).

reviews of current knowledge in this area are available (see, for example, Slinn 1983).

The most important attachment process under most in-cloud conditions is undoubtedly nucleation. Nucleation is a kinetic process in which water molecules condense from the vapor phase onto a suitable surface. Dust and pollutant aerosol particles provide such surfaces in the air. The result is a cloud of droplets (or ice crystals) containing the pollutant. The droplets may grow by the same process (condensation) or may lose water by evaporation.

The tendency of water vapor to condense on aerosol particles depends on the characteristics of the particles and the degree of saturation of the air with water vapor. As a consequence the aerosol and associated cloud particles compete for water molecules. Some particles will capture water with high efficiency and grow substantially in size. Others will acquire only small amounts of water, whereas still others will remain essentially "dry" elements. In addition, some particles may be effective for nucleation of ice crystals, whereas others will be active only for the formation of liquid water. The nucleating capability of a particular aerosol particle is determined by its size, its morphological characteristics, and its chemical composition. Acid-forming particles, by their very nature, are chemically competitive for water vapor and thus tend to participate actively as condensation nuclei for liquid water. This attribute enhances their propensity to become scavenged early in storms and has a significant effect on the nature of the acid-precipitation formation process.

Diffusional attachment, as its name implies, results from diffusion of the pollutant molecule or particle through the air to the surface of a water droplet. The process may be effective in the case of both suspended cloud elements and falling hydrometeors. It depends chiefly on the magnitude of the molecular (or Brownian) diffusivity of the pollutant; because diffusivity is inversely related to particle size, this mechanism is less important for larger particles. For practical purposes, diffusional attachment can be ignored for particles with radii of more than a few tenths of a micrometer.

The motion of a molecule or particle to the surface of a water droplet by diffusion depends on the gradient in the concentration of the pollutant in the vicinity of the surface. Thus, if the cloud or precipitation droplet can

accommodate the influx of pollutant readily (for example, the pollutant is highly soluble in water), it will effectively depopulate the adjacent air, thus making a steep concentration gradient and encouraging further diffusion to the droplet. If for some reason (such as particle "bounce off" or low gas solubility) the droplet cannot accommodate the pollutant, further diffusion to the droplet will be discouraged. If the cloud or precipitation droplet supplies the pollutant to the local air through an outgassing mechanism, the concentration gradient will be reversed and diffusion will carry the pollutant away from the droplet. In general, diffusional attachment processes are sufficiently well understood to allow their mathematical description with reasonable accuracy.

Inertial attachment arises by virtue of the facts that pollution particles and scavenging droplets are constantly in motion and that both have finite volume and mass. The most important example of inertial attachment is the impaction of aerosols by falling hydrometeors. In this case, the hydrometeor falls under the influence of gravity, sweeping out a volume in space. Collisions occur between the falling hydrometeors and some aerosol particles, resulting in attachment.

The effectiveness of impaction depends on the size of both the aerosol particle and the hydrometeor; mathematical formulas exist to estimate the magnitudes of these processes. Impaction generally becomes unimportant for aerosols less than a few micrometers in size. In this context it is interesting to note that a two-stage capture mechanism can exist, in which a small aerosol first grows through nucleation to form a larger droplet that is then captured by inertial attachment. This two-stage process, called accretion, is an essential factor in the generation of precipitation in clouds and has been postulated as an important mechanism in scavenging pollutants below clouds.

A second example of inertial attachment is turbulent collision. In this case, the particles and scavenging elements, subjected to a turbulent field, collide because of dissimilar dynamic responses to velocity fluctuations in the local air. This scavenging mechanism is thought to be of secondary importance and has received comparatively little attention in the literature, although some recent theoretical analyses have suggested it to be significant for droplets and particles of specific sizes.

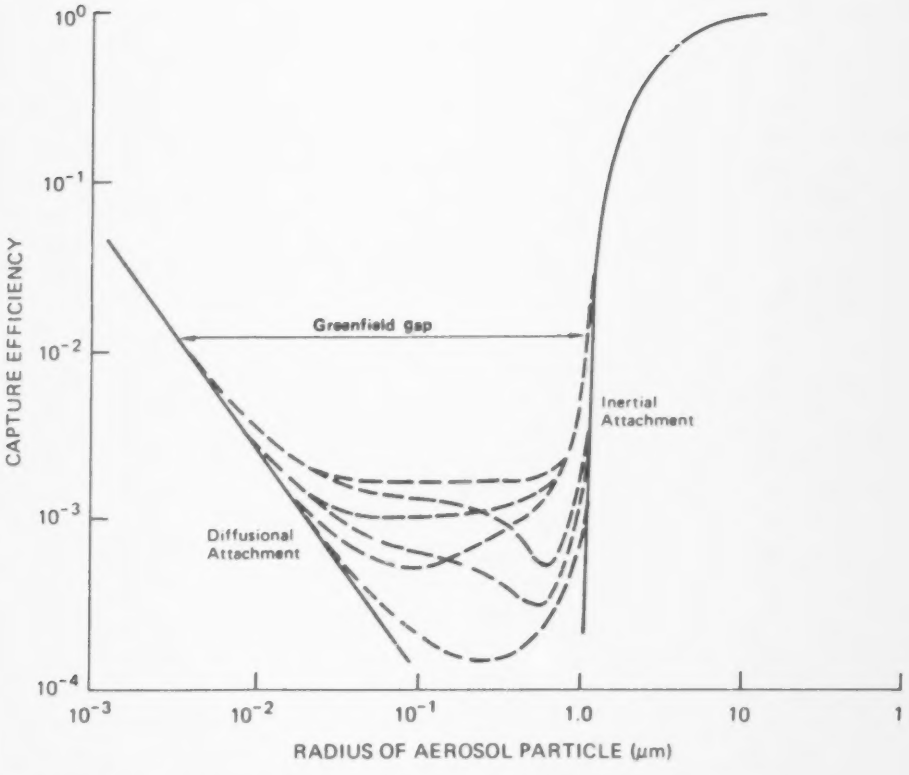

FIGURE 2.10 Theoretical scavenging efficiency of a falling raindrop of diameter 0.31 mm as a function of aerosol particle size. SOURCE: Adapted from Pruppacher and Klett (1978).

Although the diffusional and inertial attachment processes are efficient for capturing very fine and very coarse particles, respectively, neither mechanism is effective for particles in the range of 0.1 to 5 μm. The resulting minimum in capture efficiency as a function of particle size, shown schematically in Figure 2.10, is known as the Greenfield gap.

Depending on circumstances, there are several additional attachment mechanisms (including accretion via the two-stage nucleation-impaction mechanism mentioned earlier) that can operate in the Greenfield gap. The processes include turbulent deposition, electrical attraction, and phoretic effects (see Appendix C for details). As indicated by the dashed lines in Figure 2.10, these mechanisms can significantly relieve the Greenfield effect under appropriate circumstances (Appendix C).

From this discussion, it should be evident that the aggregate of possible attachment processes comprises a complex system that is difficult to characterize mathematically. This complexity, combined with the processes of formation and delivery of precipitation that occur both consecutively and concurrently, provides a major source of uncertainty in current models of regional pollution transport and deposition.

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3 Theoretical Models of Regional Air Quality

Analysis of the spatial and temporal behavior of atmospheric parameters and climatological patterns depends on a thorough theoretical understanding of the physical and chemical processes involved. That understanding, in turn, depends on observations of phenomena in the field and in the laboratory. One purpose of analyzing the relationships between emissions of precursor gases and deposition of acidic or acid-forming substances is to develop means for assessing the potential effectiveness of alternative proposals for mitigating the adverse effects of acid deposition. Uncertainties in the current understanding of the relevant physical and chemical processes are reflected in uncertainties in analytical models of these relationships.

Construction of analytical models is a typical method by which scientists approach complex problems. For many years earth scientists have been developing knowledge about flows of substances in the environment (within and among the atmosphere, hydrosphere, biosphere, and lithosphere). All elements cycle naturally through the environment; sulfur and nitrogen are two prominent examples. Models have been developed--some conceptual, some empirical, some theoretical--to organize that knowledge in ways that allow predictions to be made that are subject to testing. In recent years, this analytical approach has taken on considerable practical importance, because of the need to assess the implications of anthropogenic disturbances on natural ecological processes. So it is with models of acid deposition.

In this report we are concerned with only part of the phenomenon of acid deposition: the relationships between emissions and deposition. Models of the cycling of sub-

stances in the hydrosphere, biosphere, and lithosphere are beyond the scope of this report.

Models of the distribution of emissions through the atmosphere and their subsequent deposition can be divided into two classes: theoretical and empirical. Empirical models consist of analyses of observations in the field; Chapter 4 deals with empirical approaches used to manipulate data and test hypotheses. In the class of theoretical models are both deterministic calculations and estimates of material balance (or budgets); the current state of the art in these approaches is described below.

MATERIAL BALANCE

The method of material balance or budgeting involves assessing the gross flows of a substance into and out of compartments of the environment. The compartments are defined for the purposes of analysis; they are generally large, so that detailed behavior of constituents is not considered. Leaving out the detail, of course, means that the results may provide only general guidance and understanding.

The most straightforward approach to budgets for acid deposition is to segment processes into one or more compartments, allowing flow between the compartments (e.g., Charlson et al. 1978). Budgets for sulfur in the atmosphere have been constructed for the global atmosphere (Granat 1976) and for regions of Europe and eastern North America (e.g., Galloway and Whelpdale 1980, Granat et al. 1976, Shinn and Lynn 1979). A summary of two budgets for eastern North America is shown in Table 3.1; these calculations were made for each category by somewhat different means. They present a qualitatively similar (but quantitatively different) picture of the sulfur oxide transport and deposition in the eastern United States as well as export to the Atlantic Ocean. Other than giving estimates for the average annual deposition over large areas, these types of calculations reveal little about the consequences of changing anthropogenic emissions of sulfur or nitrogen. They also provide no guidance about the deposition of acid-producing material on specific regions that are ecologically sensitive. They do, however, provide a sense of the scale of exports of atmospheric pollutants from one region to another.

TABLE 3.1 Comparison of Atmospheric Sulfur Budget Estimates for the Eastern United States[a] and Northeastern United States[b] in teragrams (million metric tonnes) per year

	Galloway and Whelpdale (1980)[a]	Shinn and Lynn (1979)[b]
Input		
Man-made emissions	14	7.5
Natural emissions	0.5	0.6
Inflow from oceans	0.2	—
Inflow from west	0.4	—
Transboundary flow	0.7	—
	15.8	8.1
Output		
Transboundary flow	2.0	(1.1)
Wet deposition	2.5	1.5
Dry deposition	3.3	2.5
Outflow to oceans	3.9	3.0
	11.7	8.1

[a]Area east of 92° W (Mississippi River).
[b]Connecticut, Delaware, Illinois, Indiana, Kentucky, Maryland, Massachusetts, Michigan, New Jersey, New York, Ohio, Pennsylvania, Rhode Island, Virginia, and West Virginia.

One application of the method has been to assess the transport of pollutants across international boundaries. Because certain pollutants, particularly sulfates and nitrates, may be transported large distances from the sources of their precursor gases, air pollution is an interstate and even an international issue. Not all the sulfur and nitrogen emitted from sources in the United States comes to the ground in the United States, and not all the sulfur and nitrogen that comes to the ground in the United States is emitted from sources in the United States. The same, of course, can be said for states and regions within the United States.

It has been estimated that, of the total sulfur emitted to the atmosphere in the eastern part of the United States, about one third is transported to the western Atlantic Ocean and beyond, while roughly one sixth is exported to Canada. The remainder, about one half, falls in the United States (Galloway and Whelpdale 1980).

The fraction of the exports of atmospheric sulfur from the United States to Canada that is deposited in Canada is unknown. It has been hypothesized that the fraction of Canadian emissions of sulfur that falls in Canada is larger than the fraction of U.S. exports to Canada that

falls in Canada. This supposition can be explained by the differences in the deposition processes for SO_2 and sulfates and the fact that U.S. exports of atmospheric sulfur to Canada are likely to be richer in sulfates than Canadian emissions. Nevertheless, more sulfur is deposited than emitted in eastern Canada (Galloway and Whelpdale 1980), so U.S. exports can account for substantial quantities of the sulfur deposited there.

DETERMINISTIC MODELS

Most of the effort to develop models of acid deposition during the past decade has been devoted to deterministic descriptions of the distribution of sulfur oxides in plumes. The work has grown from efforts to develop plume models for studying effects of emissions on ambient concentrations of pollutants at relatively small distances from sources. Current models used to analyze regional pollution problems such as acid deposition apply to areas of the order of 10^6 km^2 and focus on long-term (annual) average behavior, taking into account emissions, airflow, mixing, chemical transformations, and both wet and dry deposition. Generally, chemical transformations and deposition processes are treated parametrically, whereas transport is calculated using available data on wind fields, for example. The models are based on sets of continuity equations for concentrations of the species of interest; the continuity equations are coupled through terms representing the production and destruction of species in chemical reactions. The equations are solved using computers.

In effect, deterministic models represent detailed material balance calculations analogous to the compartmentalization approach mentioned earlier, but in this case the compartments in the atmosphere are much smaller, so detailed behavior must be included.

Once confidence in deterministic models has been achieved, through testing and verifying, it should be possible to use them to assess the potential consequences of alternative proposals for mitigating acid deposition, since sensitivity tests would be feasible with this type of model.

There is a variety of regional models for average deposition rates of sulfur oxides over eastern North America (e.g., U.S./Canada Work Group #2 1982). The models use different approximations to characterize

atmospheric processes (Table 3.2). They have not been verified systematically because of a lack of observational data. However, testing and initial comparisons of several models for annual averages indicate that their accuracy in estimating either ambient SO_x concentrations or wet-deposition rates is inadequate for quantitative assessment of the effects of emissions from specific sources (U.S./Canada Work Group #2 1982). Initial comparisons show no preference by performance for a specific model for application to the situation in eastern North America, although from the limited number of comparisons currently available, it appears as if models that treat meteorological parameters in a gross statistical sense appear to perform as well as the more sophisticated models (U.S./Canada Work Group #2 1982).

At least three models (SURADS, RTM-II, and STEM) are capable of simulating regional sulfate pollution episodes over eastern North America (Table 3.2). These models use added sophistication in treating atmospheric processes, including incorporating multilevel winds and mixing, diurnally varying chemistry according to photochemical modeling, and variable dry-deposition rates. However, the SURADS model has not incorporated cloud processes and wet deposition in published applications. Tests of the SURADS model against the data from the Sulfate Regional Experiment yielded promising results for ambient sulfate conditions but less satisfactory results for sulfur dioxide concentrations (Mueller and Hidy 1982a). The other two models, RTM-II and STEM, incorporate cloud processes and other aspects of precipitation chemistry, but their performance in comparison with observations has not been reported.

Treatment of Transport and Mixing

Because long-range transport is at the heart of the controversies surrounding acid deposition, we review here the ways in which regional-scale models typically treat trajectory analysis.

Meteorologists have approached the transport problem in a number of ways. The simplest method is to use observed values of horizontal winds at specified altitudes to calculate by interpolation where the winds would carry a given air parcel containing the material of interest (i.e., Lagrangian or trajectory model). This type of trajectory model has been widely used and is referred to

TABLE 3.2 Characteristics of Several Regional-Scale Air Pollution Models Used to Assess Acid Deposition

Characteristic	AES[a]	ASTRAP[a]	CAPITA[a]	ENAMAP-1[a]	MEP[a]	MOE[a]
Type	Lagrangian–box	Statistical–trajectory	Monte Carlo	Puff–trajectory	Lagrangian	Statistical
Output	Monthly SO_2 and $SO_4^=$ concentrations and dry and wet S depositions	Monthly SO_2 and $SO_4^=$ concentrations and dry depositions and bulk S wet deposition	Monthly SO_2 and $SO_4^=$ concentrations and dry and wet depositions	Monthly SO_2 and $SO_4^=$ concentrations and dry and wet depositions	Monthly concentration and dry and wet depositions of sulfur	Long-term SO_2 and $SO_4^=$ concentrations; annual dry and wet sulfur depositions
Input	Annual and seasonal SO_2 emissions; daily precipitation amounts; winds and temperatures twice daily at four heights	Annual and seasonal SO_2 emissions for six-layered grid; stack parameters for major sources; 6-h precipitation amounts; wind profiles twice daily	Annual SO_2 emissions; 6-h precipitation probabilities; twice daily upper-air wind profiles and three times daily surface winds	Annual SO_2 emissions; 3-h precipitation amounts; wind profiles twice daily	Annual and seasonal SO_2 emissions; 3-h precipitation amounts; 6-h surface pressures	Point sources; area sources treated as effective point sources; statistics of durations of wet and dry periods and average precipitation rate during wet periods; statistical treatment of winds
Number of cells in the grid	52 × 37	User specified	52 × 37	46 × 41	User specified	User specified
Grid size (km)	127 × 127	Receptor point locations	127 × 127	70 × 70	Point receptors	Point receptors
Analysis of precipitation (by preprocessor)	Objective analysis of daily precipitation amounts	Hourly data are summed to produce 6-h totals across a grid of about a 76-km spacing	No actual precipitation rates used; time averages of precipitation probabilities used for each grid square	Hourly U.S. and 6-h Canadian data are summed to produce 3-h totals	Objective analysis of 3-h amounts	Climatological lengths of Eulerian and Lagrangian wet and dry periods; rate of 1 mm/h

Mixed layer	Monthly climatological heights	Diurnal pattern including nocturnal surface-based inversion, maximum 1000 (w) and 1800 (s)	Day: 800 (winter), 1200 (spring/fall), 1350 (summer), Night: 300	1150 (winter), 1300 (spring/fall), 1450 (summer)	Varies diurnally over model domain	1000
Oxidation rate for SO_2 (percent/h)	1.0	Varies diurnally: 0.2-5.5 (summer), avg. of 2.0; 0.1-1.5 (winter), avg. of 0.5	0.6 (winter), 1.2 (summer)	1.0	Seasonal and diurnal variation: mean of 1.0	1.0
Dry deposition velocity (cm/sec)					Seasonal and diurnal variation: mean of	
SO_2	0.5	Vary diurnally: 0.45 (summer avg.), 0.25 (winter avg.) (SO_2 and SO_4^{2-} similar but not identical)	0.31 (winter), 1.20 (summer)	0.38 (winter), 0.48 (summer)	0.75	0.5
SO_4^{2-}	0.1		0.07 (winter), 0.15 (summer)	0.22 (winter), 0.28 (summer)	0.25	0.05

TABLE 3.2 (Continued)

Characteristic	AES[a]	ASTRAP[a]	CAPITA[a]	ENAMAP-1[a]	MEP[a]	MOE[a]
Wet removal rate (percent/h)						
SO_2	$3 \times 10^5 P_{24}(t)/H$	--	0.6 PP (winter); 11.7 PP (summer)	28.0 $P_1(t)$	Dependent on pH and temperature	10.8
$SO_4^=$	$85 \times 10^5 P_{24}(t)/H$ where H is the mixed layer (mm)	--	5.0 PP (winter); 29.0 PP (summer) where PP = probability of precipitation (%) each 6-h period	7.0 $P_1(t)$	Precipitation rate dependent: mean of 30 $P_1(t)^{0.5}$	36.0 (used in stochastic scavenging model with $\tau_d = 56$ h and $\tau_w = 7$ h (the avg. dry- and wet-period durations)
Bulk S	--	Minimum of $100(P_6(t)/10)^{1/2}$ and 80	--	--	--	--

$P_1(t)$ = liquid precipitation rate (mm/h)

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TABLE 3.2 (Continued, Part 2)

Characteristic	RCDM-3[a]	UMACID[a]	ELSTAR[b]	RTM-II[c]	STEM-I[d]	STEM-II[e]	SURADS[f]
Type	Analytical	Puff-trajectory	Lagrangian trajectory	Hybrid Lagrangian/ Eulerian	Time-dependent Eulerian	Time-dependent Eulerian	Eulerian
Output	Monthly concentration and dry and wet deposition of sulfur	Estimates of source contributions to downwind concentrations and contributions of upwind sources on receptors at 6-h time steps	Hourly concentrations of photochemical products and intermediates; dry deposition; budgets for SO_x, NO_x	3-h, daily, monthly concentrations of SO_2, $SO_4^=$, wet and dry sulfur deposition	Hourly and daily SO_2 and $SO_4^=$ concentrations; instantaneous and accumulated SO_2 and $SO_4^=$ deposition; daily regional sulfur budget	Hourly and daily species concentrations; instantaneous and accumulated deposition rates	Hourly and daily SO_2, $SO_4^=$ concentrations; dry deposition rates; daily SO_2 budget
Input	Emissions and centroids of emissions for area sources, spatially averaged annual and seasonal wet and dry periods; monthly, seasonal, and annual resultant winds and persistence factors	Annual SO_2 emission rates; 3-h precipitation amounts; wind profiles twice daily	Emissions of SO_x, NO_x, HC; winds extrapolated hourly in all layers from surface conditions, temperature profiles	Daily or hourly emissions; meteorological data	Surface and upper-air winds, cloud cover and ceiling, surface roughness, evaporation rates, vertical temperature and humidity profiles	Surface and upper-air winds, cloud cover and ceiling, rainfall rate, surface roughness, evaporation rates, vertical temperature and humidity profiles	Initial SO_2 and sulfate updated hourly; hourly SO_2 emissions; estimates of NO_x and HC emissions; radar-based precipitation; 12-h synoptic wind field aloft at fine levels; hourly surface winds; temperature profile, sunlight, cloud cover
Number of cells in the grid	70 × 70	41 × 32	Variable: 100 × 100	52 × 46 × 2 (episodic), 26 × 23 × 2 (long term)	User specified	User specified	Variable: e.g., 20-23
Grid size (km)	80 × 80	80 × 80	5 × 5	40 × 40 (episodic), 80 × 80 (long term)	80 × 80	Regional scale	80 × 80
Analysis of precipitation (by preprocessor)	Spatially averaged precipitation amounts and the average durations of wet and dry periods	Hourly data summed for 3-h periods for each 80-km grid square	Not included	Objective analysis on hourly data to generate grid-by-grid rates	— —	— —	Cloud chemistry and scavenging algorithm in fine layers

TABLE 3.2 (Continued)

Characteristic	RCDM-3[a]	UMACID[a]	ELSTAR[b]	RTM-II[c]	STEM-I[d]	STEM-II[c]	SURADS
Mixed layer	600 (winter), 1200 (summer), 1000 (annual)	Varies only with month	4 or 5 layers as function of maximum mixing height on test day	Linear interpolation of daily minimum and maximum; spatial interpolation by spline fit	Calculated, varies diurnally	Calculated, varies diurnally	Five layers to 1500 m, preprocessors calculate mass conservative winds by layer and estimate diurnally variable mixing in each layer
Oxidation rate for SO_2 (percent/h)	Chemical conversion time scale 2.4×10^5 sec (1 percent/h)	Winter: 1.4 (day); 0.1 (night); Summer 2.8 (day); 0.2 (night)	Variable; tied to photochemical model	Function of solar zenith angle and geographic location northeastern U.S. in summer, varies from 0.1 to 2.0 with 0.78 avg.; northern Great Plains, 0.3 avg.	– –	– –	Constant (0.5 to 3.0) or diurnally variable as calculated
Dry deposition velocity (cm/sec)	Weighted by the percent of dry time	Varies according to time after sunrise and land use:					
SO_2	0.50	0.10-0.55 (winter); 0.10-0.82 (summer)	Varies with aerodynamic resistance	One-dimension diffusional model; depends on land use and time of day	Calculated based on surface type, evaporation, and meteorological conditions	Calculated based on surface type, evaporation, and meteorological conditions	Diurnally variable, calculated
$SO_4^=$	0.05	0.05-0.28 (winter); 0.03-0.43 (summer)	0.55 times SO_2 rate				0.1 to 1.0 times SO_2 rate

Wet removal rate (percent/h) SO_2	--	$5.0 \times 10^{4} P_x(t)/H$	Not included	Depends on precipitation rate and cloud type	User-specified subroutine to calculate zeroth and first-order removal rate constants	Calculated based on liquid-water content of clouds, temperature, pH, rainfall intensity, drop size and number distribution, and chemical and physical properties of the absorbed species	Not included
$SO_4^=$	--	$2.32 \times 10^{5} P_x(t)^{0.625}/H$ where H = mixing height (mm)					
Bulk S	$\Lambda P_i \frac{T_d + T_w}{T_w}$, where T_d and T_w are the average duration of the dry and wet periods (h), respectively, and Λ is the scavenging coefficient equal to 0.34	--					

$P_x(t)$ = liquid precipitation rate (mm/xh)

[a]U.S./Canada Work Group #2 (1982).
[b]Lloyd et al. (1979).
[c]Durran et al. (1979); Liu et al. (1982), and Stewart et al. (1983).
[d]Carmichael and Peters (1983).
[e]Carmichael et al. (1983).
[f]Mueller and Hidy (1982b).

as a constant-level (isobaric) trajectory (Heffter and Ferber 1977). The vertical component of the wind is ignored in this approach, so the method does not give true trajectories in many circumstances, such as near frontal zones or over a mountain range.

A more sophisticated, but not necessarily better, approach is to assume an isentropic trajectory. In essence this method takes account of the fact that an air parcel will not in all cases follow a horizontal pressure surface but will remain on a surface of constant potential temperature. Calculations of isentropic trajectories therefore incorporate some large-scale vertical motions. However, the isentropic model still does not account for vertical motions in clouds or when isentropic surfaces intersect the terrain. The more complicated the single-trajectory models become, the less application they have in day-to-day operational use because of increased requirements for computer time and power.

Single forward trajectories are rarely used to investigate precipitation chemistry because many sources usually contribute acidic precursors along the path of flow. An alternative approach is to compare the single backward trajectory to a given precipitation event. In back-trajectory analysis a parcel is traced back in time from a given precipitation event, allowing evaluation of the contributions of possible sources along the path of the parcel.

In the absence of multiple sources along a path, single-trajectory analysis can be useful. For example, the episodic data on precipitation chemistry collected in Bermuda have been analyzed with the technique by Jickells et al. (1982). They found that when airflow was from the North American continent the acidity of the precipitation was an order of magnitude higher than when the rains were associated with flows from other directions. The North American continent evidently is the major source of acidic precursors in Bermuda, which is more than 1,000 km from the mainland.

Limitations of single-trajectory analysis in describing transport have caused meteorologists to develop multiple-trajectory Lagrangian models and models based on calculations at fixed points in space. In the latter (Eulerian) model, concentrations of atmospheric constituents at fixed points are calculated for successive intervals of time based on concentrations calculated for previous intervals., Because the fixed points in space

are usually arranged in a grid pattern, the model is also known as a grid model.

Single-trajectory calculations directed backward from receptors are sometimes helpful for identifying source areas frequently contributing to deposition at selected receptor sites. Grid models are most efficiently used for weighting the contributions of many sources to several receptors.

Early work comparing back trajectories to limited event data on precipitation chemistry indicated that in the Northeast the more acidic precipitation came with flows from the Southwest (Kurtz and Schneider 1981, Miller et al. 1978). More recent studies using an improved data base show that single-trajectory analysis at a site in the Northeast suggests a regional acid precipitation phenomenon (Wilson et al. 1982).

Compared with the interior of the United States, a much larger portion of the average annual precipitation in the Northeast is associated with cyclonic disturbances (low-pressure systems). The main tracks of cyclonic storms tend to converge in the northeastern United States and southeastern Canada (see Figure 4.7, Chapter 4). Thus the areas of eastern North America regarded as being particularly sensitive to acid deposition receive precipitation from storm systems with origins at almost any latitude in the interior or the West. Additional evidence that the air shed of the sensitive area is of synoptic scale is presented in Chapter 4. To date, acid-deposition models treat transport and mixing in terms of trajectories and diffusion. Episode models in current use take account of air pollution embedded in synoptic-scale weather systems, but they require great detail in inputs on wind, temperature, and moisture fields.

Verification of any model is critical to its acceptance both as a description of scientific understanding and as a tool for analyzing policy choices. Methods are available for testing the validity of trajectory calculations. Among the first was the use of special balloons, called tetroons, that maintain a constant altitude (Pack et al. 1978). Tetroons were released and tracked on the assumption that they behave like an idealized air parcel, despite the fact that they are confined to a surface of constant density. Because of logistic and other problems, however, this method has all but been abandoned, except in short-range experiments. A more effective method of

verifying trajectory calculations is to release an inert tracer, such as perfluorocarbon or an isotopically rare methane (such as CD_4), that can be measured 1,000 km downwind or more. Plans are now being made to release tracers under a number of meteorological conditions to evaluate trajectory calculations.

While techniques for computing trajectories have advanced markedly over the past decade, they still are plagued by a host of uncertainties. One difficulty that all such techniques share is the sparsity of meteorological data. Upper-air soundings are made only twice daily at widely separated stations in the United States.

More fundamentally, trajectory models incorporate simplifying assumptions, such as isobaric or isentropic behavior, that only approximate the true behavior of air masses. Consequently, in situations in which the assumptions are not valid, model calculations may be unreliable. An example is the vicinity of storm systems, in which complex flows and thermodynamic processes limit severely the validity of the single-layered approach.

Two different calculations of a trajectory in the vicinity of a frontal storm system appear in Figure 2.3, Chapter 2. One uses a constant-pressure (isobaric) surface and the other a constant-entropy (isentropic) surface (Davis and Wendell 1977). The trajectories were calculated for the same conditions and start from the same point (near St. Louis). The calculated positions of the two theoretical air parcels after 24 hours of transport are several hundred kilometers apart. Although the isentropic trajectory is expected to be closer to reality, Figure 2.3 contains no information about the location of the "real" trajectory in this case. The plot provides a graphic illustration of the level of uncertainty in current capabilities for predicting trajectories.

The weaknesses in trajectory models are of particular concern because it is precisely in storm environments--for which current models are weakest--that it is most important to trace pollutants with accuracy. Although transport theory suggests that errors in estimates of trajectories in clear air will tend to cancel if averages are taken over multiple events, large systematic biases may be expected in storm situations. Such uncertainties make attribution of specific deposition events to specific sources particularly difficult.

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Treatment of Transformation Chemistry

The extent to which realistic chemistry is incorporated into current regional models varies significantly. Most models used recently (see Table 3.2) employ absolutely no chemistry but include instead only a fixed SO_2 transformation rate (usually 1 to 4 percent/h). Results from such models should not be relied on in the development of control strategies for regional air-quality problems in which chemical phenomena play a central role.

Attempts have been made by Rodhe et al. (1981) to include some chemistry (19 reactions) in their transformation-transport model. However, this work includes only a rudimentary scheme of gas-phase reactions involving only molecules of O_3, CO, CH_4, C_2H_4, CH_2O, H_2O_2, NO, NO_2, HNO_3, and H_2SO_4 with intermediates of HO, $O(^1D)$, $O(^3P)$, and HO_2. One aqueous-phase reaction was included superficially as $SO_2 + H_2O_2 + \text{"CLOUD"} \rightarrow H_2SO_4$.

It is interesting, however, that Rodhe et al. concluded (p. 139) that "When dealing with a coupled chemical system like the one we have studied one may not assume a proportional dependence of concentrations on emission rates." They suggest, as we do, that models must include the essential chemistry that controls the oxidation of SO_2, NO, NO_2, and hydrocarbons in the atmosphere. It is almost certainly true that the scheme of Rodhe et al. fails to do this properly.

Other chemical reaction schemes have been adapted for regional-scale models. Mueller and Hidy (1982b) have reported the application of a homogeneous gas-phase reaction scheme in the SURADS model. This calculation is run to generate a diurnally varying oxidation rate for sulfate that accounts for hydrocarbon vapor/nitrogen oxide emissions and their atmospheric distribution. This model currently is being extended to incorporate cloud chemistry in the chemical processing. It has been used in an integrated stepwise manner in the ELSTAR trajectory model (e.g., Lloyd et al. 1979). The STEM series of models (e.g., Carmichael et al. 1983) and the air shed model reported by Stewart et al. (1983) incorporate a significant number of gas-phase reactions and cloud processes into an Eulerian grid calculation. The performance of the chemistry in these very complex numerical simulation schemes has not yet been compared with simpler schemes or with each other.

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Treatment of Dry and Wet Deposition

Dry deposition is usually treated parametrically in models rather than in terms of the fundamental physics of the processes. For example, simple treatments assume that the rate at which gaseous SO_2 or sulfate aerosol particles reach the ground is directly proportional to the atmospheric concentrations of the respective pollutants near the ground. The constant of proportionality, called the deposition velocity, is often assumed for simplicity to be independent of spatial coordinates and time. Because dry-deposition processes depend on characteristics of ground cover, surface roughness, and stability of the air layer immediately above the surface, more sophisticated treatments use variable deposition velocities according to the spatial dependence of these conditions. The deposition velocity may also be taken to vary diurnally or can be estimated from aerodynamic parameters, such as surface winds. Resistance to gas-phase transfers associated with the stomata of leaves or the assimilation capacity of underlying surfaces can also be incorporated in theoretical treatments. The dry-deposition velocities for sulfate aerosol particles are usually assumed to be somewhat less (between 10 and 50 percent) than those for gaseous SO_2. More generally, the rate of particle deposition is dependent on size, but currently available models have not yet taken this dependence into account. Dry deposition of NO_2 differs somewhat from that of SO_2 and is sometimes taken to be as little as one half that of SO_2.

There are no regionally representative measurements of dry deposition. Therefore the parameterizations adopted in models have not been tested extensively. Instead they are based on limited experiments in the field or in wind tunnels. Thus the limitations of the parameterizations have not been established quantitatively. On the basis of comparison of model outputs with measurements, parameterization of dry deposition should be known within a factor of 2, but the range of measurements is much larger than this (e.g., Sehmel 1980).

The parameterization of wet deposition is extremely difficult for episode calculations because of the variability in time and space of cloud cover, cloud depth, and precipitation. Over a long period of time, such as a year, averaged wet deposition is assumed to be proportional to the total quantity of precipitation or

the precipitation rate. The parameterization of wet deposition typically is one of two types, both of which should be considered as being in rudimentary stages of development.

The first class of parameterization involves the scavenging coefficient, which is defined as the rate at which an air pollutant is incorporated into precipitation per unit volume divided by the local concentration of the pollutant. If the scavenging coefficient is known, then wet removal can be calculated in a relatively straightforward manner on the basis of airborne pollutant concentrations. Although in principle the scavenging coefficient varies in space and time, the usual practice is to use "storm-average" values.

The second class of parameterization involves the scavenging ratio, which is the ratio of the concentration of the pollutant in precipitation at ground level to the concentration of the pollutant in air at ground level. The scavenging ratio is somewhat easier to apply in practice than the scavenging coefficient because the former does not require a vertical integration of pollutant concentration to obtain the deposition rate.

Both the scavenging coefficient and the scavenging ratio have well-established theoretical bases. Considerable theoretical work has been applied to their physical evaluation. The parameterizations suffer from the difficulty, however, that they consolidate the effects of a large number of complex physical processes. As a consequence the parameters conceal a great deal of uncertainty, particularly when aqueous-phase transformation processes are involved. The parameters actually used in regional models tend to be based more on empirical data than on analysis of actual mechanistic behavior. The proportionality coefficients are often deduced from comparison of ambient air concentrations and precipitation chemistry or from semiempirical models (e.g., Scott 1978, 1981). Recent diagnostic models of scavenging appear to be moderately successful in resolving some of these individual mechanisms, and these techniques can be expected to be applied to regional models in the future.

Considering the crudeness of parameterization of wet and dry deposition, it is surprising that the linear models perform better in estimating the long-term average wet deposition of sulfate than in estimating the ambient concentrations.

Linearity or Nonlinearity in Theoretical Models

Physical Processes

A deterministic source-receptor model generally provides the solutions (or approximations to the solutions) of a set of conservation equations for each species of the form

$$\frac{\partial c_i}{\partial t} = -\vec{\nabla} \cdot c_i \vec{v} - w_i + r_i .$$

The relationship, called the continuity equation, simply expresses a material balance for pollutant species i in terms of the time rate of change of its time-averaged concentration (c_i), the flow and diffusion field ($\vec{\nabla} \cdot c_i \vec{v}_i$), its rate of removal by deposition processes (w_i), and its net rate of production by chemical reaction (r_i). Each of the parameters in the equation in general varies as a function of position $\vec{x}$ and time. This equation, together with suitable boundary and initial conditions, is considered to be a complete mathematical description of any constituent in the air.

A profusion of methods has been applied for solving equations of this type, and a correspondingly large number of "models" for solutions exists. The characteristics of a number of models are given in Table 3.2. A comprehensive discussion of the methods of solution used in these models is beyond the scope of this report; it is important, however, to recognize that, whatever the class to which a particular model belongs, the model still may be described in terms of the continuity equation or its derivatives and may be viewed as a solution to these equations with appropriate initial and boundary conditions.

Recently, in connection with proposals for mitigating acid deposition, the question has arisen as to whether deposition rates are linear functions of emissions. It is therefore appropriate to discriminate between linear and nonlinear models and to indicate their significant differences.

The continuity equation and its boundary conditions can be mathematically linear or nonlinear, depending on whether nonlinear operators act on the dependent variable c_i, the concentration of species i. A linear operator L is one that satisfies the relationship

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$$L(\alpha c_i + \beta c_j) = \alpha L(c_i) + \beta L(c_j),$$

where α and β are arbitrary constants. Thus, for example, if the net rate of production (r_i) of species i through chemical reactions is proportional to the first power of c_i, such as

$$r_i = \kappa_1 c_i,$$

r_i is linearly related to c_i. If, however, $r_i = \kappa_2 c_i^2$, the relationship is nonlinear.

While detailed discussion of the continuity equation and its solutions is beyond our current purpose, we note that solutions to linear and nonlinear forms of the continuity equation have characteristics that are of practical importance in formulating control strategies. An important feature of linear systems of this type is that the results satisfy the principle of superposition. From the characteristics of linear operations we can find the combined consequences of, say, two sources simply by summing the contributions of each calculated without regard for the other. This practice is central to Lagrangian trajectory modeling and is not applicable if nonlinear processes predominate.

Let us suppose, for example, that a simple form provides the following relationship between the magnitude of the pollution source S_{ij} at location $\vec{x}_j$ and a resulting ambient concentration at a specified location $\vec{x}$:

$$c_i(\vec{x},t) = k_{ij} S_{ij}(\vec{x}_j,t).$$

The coefficient k_{ij} depends on the separation of the source and receptor locations $(\vec{x} - \vec{x}_j)$ and is assumed to be constant. The concentration c_i is proportional to the first power of S_{ij}, which is to say that c_i is linearly related to S_{ij}. For the linear form of this type, a change in S_{ij} would result in a proportionate change in c_i. For example, a 50 percent reduction in S_{ij} would result in a 50 percent reduction in c_i.

Simple linearity does not, however, guarantee proportionate (one-for-one) reductions. Suppose, for example, that there are a number of sources (N, for example) influencing c_i and that a substantial background concentration B_i (due to natural sources) is present. In this more general case, c_i has the form

$$C_i(\vec{x},t) = \sum_{j=1}^{N} k_{ij} S_{ij} + B_i(\vec{x},t),$$

where the symbol $\sum$ indicates a sum of the contributions from each of the N sources. This result is still linear in the jth source strength, S_{ij}, but if B_i and the contributions from the other N - 1 sources are not very small compared with $k_{ij}S_{ij}$, a decrease in S_{ij} would produce a less than proportionate change in c_i.

Although noted here in a rather simplistic sense, the linear relationship is the underlying basis for the so-called linear-rollback strategy of emission reduction.

Because of the complexity of atmospheric processes, it is unreasonable to expect that a simple linear equation with constant coefficients would provide an accurate description of source-receptor relationships. A more useful expression has the form

$$c_{ij}(\vec{x},t) = \sum_{j=1}^{N} p_{ij}S_{ij} + B_i(\vec{x},t),$$

where the parameter p depends on atmospheric variables, such as wind speed and rainfall rate, and changes in time and space appropriately to reflect the dependence of the atmospheric variables on time and space. Most regional source-receptor models in current use yield results of this type. Although p is functional in form, this expression is still a linear equation and necessarily stems from a linear form of the continuity equation. The implications regarding superposition and rollback discussed in the context of the simpler linear result apply here as well, provided that the functional dependence of p does not inadvertently include S.

It is in fact more realistic to expect that the function p depends explicitly or implicitly on S, for example, through the concentrations of other atmospheric species. In this case, we can write the solution to the continuity equation in the same form but with a new factor of proportionality q that depends explicitly on S:

$$c_{ij}(\vec{x},t) = \sum_{j=1}^{N} q_{ij}(S_{ij})S_{ij} + B_i(\vec{x},t),$$

where again the variables in the argument of q are functions of time and space. This result is nonlinear since S_{ij} appears both functionally in the argument of q_{ij} and as a first-order multiplier. The dependent variable c_i no longer depends on the first power of S_{ij} (the independent variable).

Because a number of atmospheric processes are nonlinear, the most accurate and complete model of the atmosphere must be nonlinear. There are computational advantages to linear mathematical systems, however, and a corresponding tendency to approximate physical processes in a manner such that linearity is obtained. Such "linearization" is a common practice in science and engineering; its success depends on the degree of deviation from linearity of the phenomenon being studied as well as on the intended application. Linearized models may do well in simulating observed regional deposition patterns, for example, whereas their capability to predict responses to specific changes in emissions may be comparatively poor.

Chemical Processes

Most researchers who have analyzed regional air-quality data have assumed a linear mechanism for transforming SO_2 into sulfate with a rate between 0.1 and 4.0 percent/h (Table 3.2). More complex reaction schemes generally have not been used to rationalize results from observations. By their nature, linear models predict a form of proportional response to emission reductions. In some cases this has previously been demonstrated to be an extremely poor assumption (e.g., in the control of photochemical oxidants in urban areas). Linearity may also be a poor assumption for circumstances involving acid precipitation. There is substantial support, however, for the argument that currently we simply do not understand the atmospheric interactions sufficiently well to supply the mathematical detail required by nonlinear concepts.

Some attempts have recently been made to incorporate chemical nonlinearities into models of acid precipitation. Most of these schemes concentrate on the production of acid sulfate as an aerosol rather than on aqueous-phase processes in clouds. As described earlier in the section on the treatment of transformation chemistry, Rodhe et al. (1981) employed a highly simplified but seemingly realistic chemical scheme for SO_2 and NO_x oxidation and found some very nonlinear effects. For example, their results indicated that a 10 percent increase in NO_x emissions would lead to a 5 percent reduction in sulfate concentration downwind from the source region, and a 10 percent decrease in SO_2 emission would result in only a 3 to 4 percent decrease in sulfate production.

Nonlinearity in the SO_2 transformation of the Rodhe et al. model was also observed in subsequent studies by Sampson (1982). He employed a somewhat improved hydrocarbon reaction scheme, but in other respects the mechanism was identical to that employed by Rodhe and his co-workers. Some of Sampson's results are reproduced in Figure 3.1. The solid lines in the figure give Sampson's results for the percentage change in ambient sulfate concentration after 24, 48, and 96 hours of transport from the source region as a function of changes in SO_2 emissions. The results of the model suggest that a relatively small reduction in sulfate levels (roughly 15 percent) may result for long transport times (96 hours) from a 50 percent reduction in SO_2 emissions.

The results of Rodhe et al. and Sampson should be treated with caution. The so-called Rodhe-Crutzen-Vanderpol model used in both studies employed specific sequences of chemical reactions and assumed uniform additions of polluted background air throughout the period of transport and transformation. Different choices of oxidation pathways and changes in the strong background source may alter the results significantly.

For example, the dashed lines of Figure 3.1 are the result of running Sampson's computer program without continuous dilution of the product mixture with background air containing sulfate (P.J. Sampson, University of Michigan, personal communication, 1982). The shift toward the linear curve (from the solid to the dashed curves in Figure 3.1) is the result of eliminating the trivial source of nonlinearity arising from the background source, term B in the equation, $c = kS + B$, considered earlier. The dashed lines of Figure 3.2 are the result of both deleting the background source of sulfate and selecting an alternative pathway for the homogeneous gas phase oxidation of SO_2. Note that the alternative assumptions give a result that is essentially linear (with proportionality constants less than unity).

The original Rodhe-Crutzen-Vanderpol model employed reaction (3.1) for oxidation of SO_2,

$$HO + SO_2 \rightarrow H_2SO_4, \qquad (3.1)$$

whereas the modification that produced the dashed curves of Figure 3.2 used

$$HO + SO_2 \ (+ O_2, H_2O) \rightarrow H_2SO_4 + HO_2. \qquad (3.2)$$

Reaction (3.1) is a single, simplified reaction in which an attempt is made to condense the chemistry that occurs in and following the primary hydroxy radical attack on SO_2:

$$HO + SO_2(+M) \rightarrow HOSO_2(+M) \qquad (3.3)$$

[Equation (A.56) in Appendix A]. See Appendix A for a more complete discussion of the reaction.

The use of reaction (3.1) is equivalent to assuming that the addition of the hydroxy radical to SO_2 terminates the chain reactions of the HO radical, and by some undefined process the initial product of reaction (3.3) leads to H_2SO_4 without regenerating a chain-carrying species. The assumption of reaction (3.1) perturbs the atmospheric reaction cycles involving HO_2 and HO radicals, which result in the oxidation of hydrocarbons, aldehydes, CO, SO_2, NO, NO_2, and other impurity species. For example, the oxidation of CO occurs in reactions (3.4) through (3.6) by way of HO-radical attack on CO:

$$HO + CO \rightarrow H + CO_2, \qquad (3.4)$$
$$H + O_2(+M) \rightarrow HO_2(+M), \qquad (3.5)$$
$$HO_2 + NO \rightarrow HO + NO_2. \qquad (3.6)$$

Note that although an HO radical is lost in reaction (3.4), another is regenerated in reaction (3.6). Similar cycles occur involving CH_2O and the hydrocarbons, for example. Now if a reaction such as (3.1) occurs, an HO radical is removed; no further regeneration of the HO radical occurs.

In writing reaction (3.2), we assume in accordance with experience in other atmospheric reaction cycles that a chain-carrying radical (HO_2) is developed following the occurrence of reaction (3.3). For example, reaction (3.2) summarizes the net result of the sequence (3.3), (3.7), and (3.8):

$$HO + SO_2(+M) \rightarrow HOSO_2(+M), \qquad (3.3)$$
$$HOSO_2 + O_2 \rightarrow HO_2 + SO_3, \qquad (3.7)$$
$$SO_3 + H_2O \rightarrow H_2SO_4. \qquad (3.8)$$

Presumably, reaction (3.7) would often be followed by regeneration of the HO radical through reaction (3.6), at least in NO-rich polluted atmospheres.

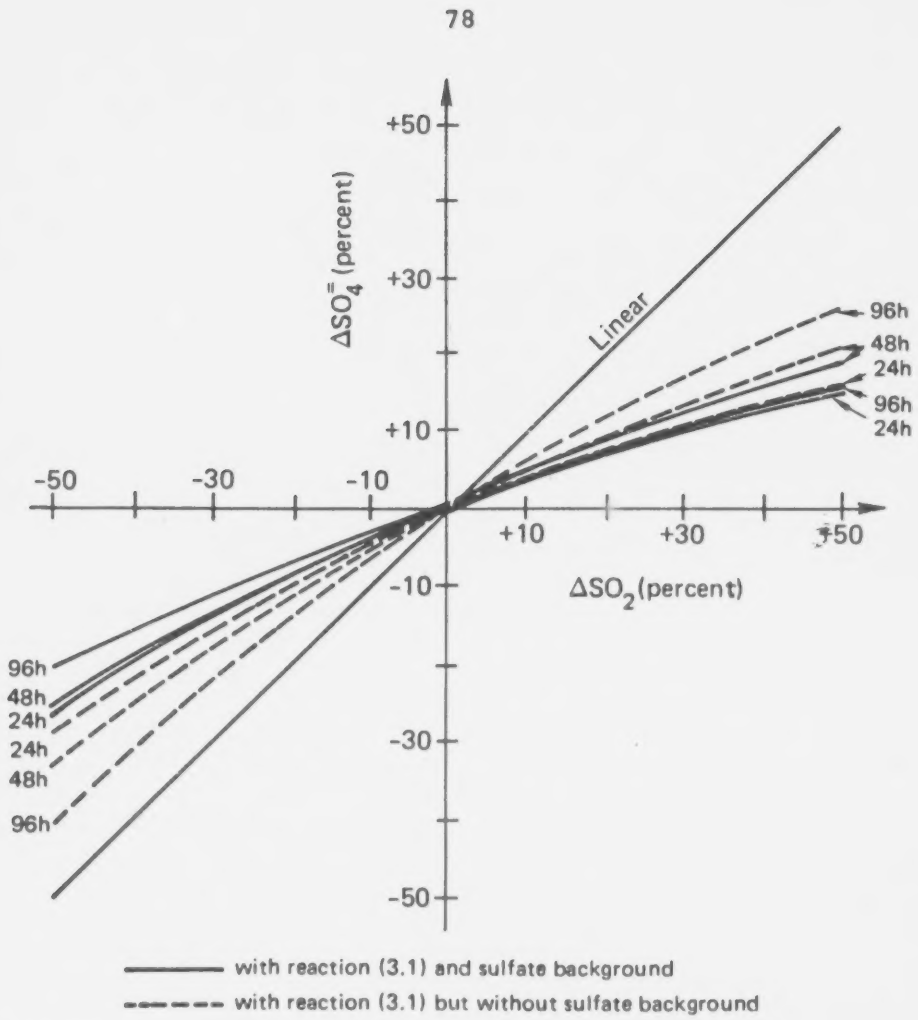

FIGURE 3.1 Effect of the assumption of background sulfate on the Rodhe-Crutzen-Vanderpol model for chemical transformation. SOURCE: Sampson (1982) and P.J. Sampson, University of Michigan, personal communication (1982).

The participation of reaction (3.1) results in a direct nonlinear feedback into the SO_2 oxidation mechanism, while reaction (3.2) does not seriously perturb the concentration of the hydroxy radical. The best available experimental evidence today supports the contention that the HO level in reacting mixtures of hydrocarbons, NO_x, and SO_2 is relatively insensitive to SO_2 concentrations and that the sequence (3.3), (3.7), (3.8), or some similar chain-propagating reactions, is important (Stockwell and Calvert 1983). In the experiment, Stockwell and Calvert varied the amount of

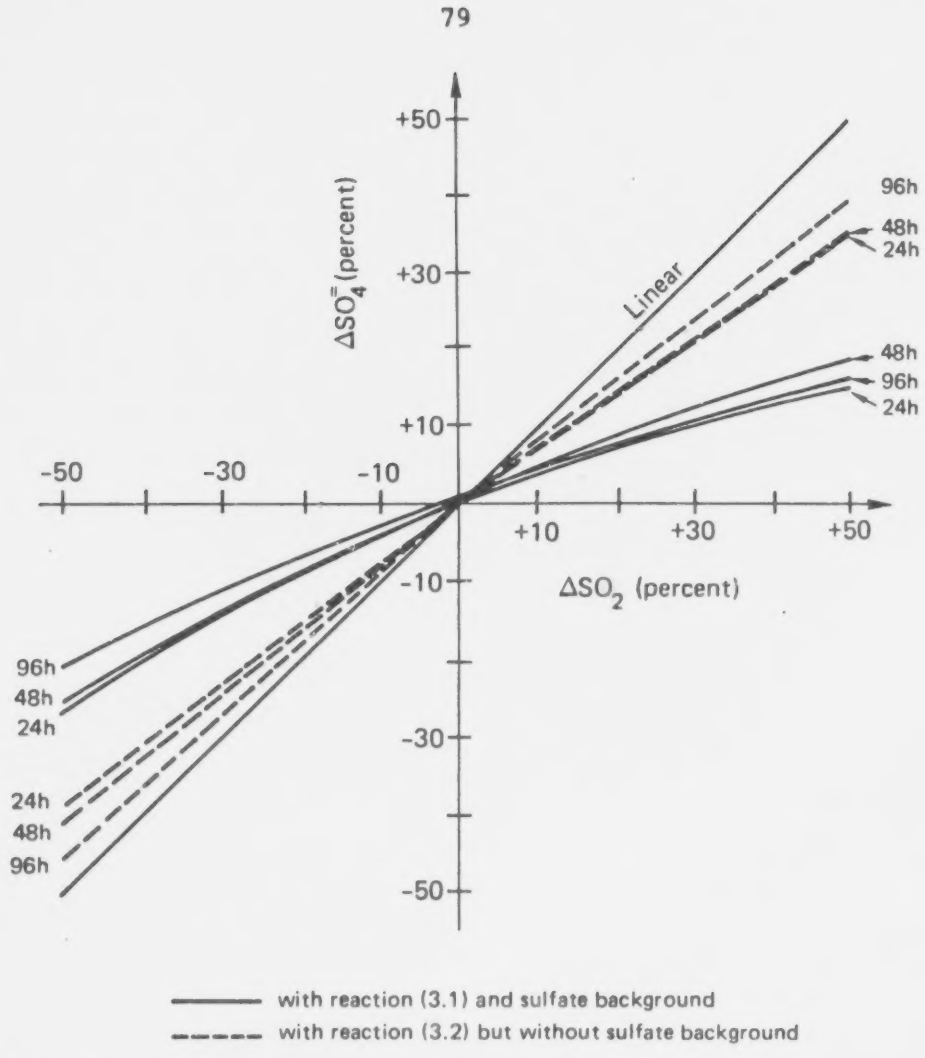

FIGURE 3.2 Effect of the assumptions of background sulfate and chain termination on the Rodhe-Crutzen-Vanderpol model for chemical transformation. SOURCE: Sampson (1982) and P.J. Sampson, University of Michigan, personal communication (1982).

SO_2 in dilute, irradiated mixtures of CO, HONO, and NO_x in air (at 1 atm), monitored the concentration of HO radicals by measuring the rate of formation of CO_2, and observed the ultimate formation of H_2SO_4 aerosol as identified by its infrared spectrum. Within the limits of experimental error, the concentration of HO radical was found to be insensitive to the concentration of SO_2 even when as much as one half of the HO radicals in the system reacted with SO_2 leading ultimately to

the formation of sulfuric acid aerosol. Chain termination as implied in reaction (3.1) was found not to be important.

The main point to recognize from this discussion is that either an apparent near linearity or a nonlinearity in the model may result from different, rather subtle, simplifying assumptions related to the choice of chemical mechanism. We conclude from these results that deviations of SO_2 conversion rates from linearity with respect to SO_2 concentration may be much smaller than has been implied recently from the results of simulations employing the seemingly realistic yet simplified reaction schemes.

Generation of nitric acid in gas-phase reactions does involve termination of an HO-radical chain directly via

$$HO + NO_2(+M) \rightarrow HNO_3(+M), \qquad (3.9)$$

and in this case we must expect the concentration of the reactant HO to be a function of the NO_2 concentration.

The concentration of the HO radical in an air mass is determined by the rates of reaction that generate it and those that destroy it. That is, at any time t the steady-state concentration of HO is given by

$$[HO] = \Sigma(R_i)_t / \Sigma k_i [A_i]_t,$$

where $\Sigma(R_i)_t$ is the sum of the rates of all HO-radical generating reactions at time t, k_i is the rate constant for the ith removal reaction of HO with reactant A_i, and the summation $\Sigma k_i [A_i]_t$ extends over all HO-loss reactions. It should be noted that reaction (3.9) is only one of several HO-HO_2-radical chain termination reactions that occur in the troposphere. Thus in theory the effect of small changes in the concentration of NO_2 on the concentration of HO is not expected to be dramatic. For example, computer simulations of the chemistry of the polluted atmosphere (see the mechanisms of Calvert and Stockwell 1983) show that only about 10 percent of the HO-HO_2-radical termination occurs through the HO-NO_2 reaction (3.9) for a tropospheric air mass typical of an urban, polluted area with an ambient concentration of NO_x of 100 ppb at sunrise. Air masses containing one tenth and one one-hundredth of this concentration of NO_x at sunrise, but the same levels of other pollutants as before, give about 0.1 and 0.01 percent of the total HO-HO_2-radical chain termination through reaction (3.9). The time dependence of the concentrations of

reactants that form HO or react to destroy it are complex functions of the initial pollutant concentrations, so that the quantitative effect of the concentration of HO on NO_x initial concentration can be obtained only through detailed calculations. However, the net effect of lowering the initial NO_x concentration by a factor of 10 (from 100 to 10 ppb) while keeping all other impurities at the same fixed level of the highly polluted air mass is to lower the maximum HO concentration from 1.6×10^{-7} to 0.96×10^{-7} ppm, only a factor of about 1.7. Clearly the dependence of HO concentration is not so sensitive to NO_x concentration as one might have expected at first consideration. Thus a more detailed analysis of the complex homogeneous chemistry of the troposphere predicts that the relationship between changes in ambient concentrations of SO_2 and changes in gas-phase formation of sulfate should exhibit only small deviations from linearity. The simple theoretical considerations of Oppenheimer (1983) lead to the same conclusion.

Nonlinear conversion of SO_2 to sulfate can in theory result from the liquid-phase oxidation of SO_2 (HSO_3^-) by hydrogen peroxide (H_2O_2). For certain atmospheric conditions a limited supply of H_2O_2 may exist in the atmosphere through gas-phase reactions (3.10) to (3.12):

$$2HO_2 \rightarrow H_2O_2 + O_2, \qquad (3.10)$$

$$HO_2 + H_2O \rightarrow HO_2 \cdot H_2O, \qquad (3.11)$$

$$HO_2 \cdot H_2O + H_2O \rightarrow H_2O_2 + H_2O + O_2. \qquad (3.12)$$

The rate of hydrogen peroxide generation in reactions (3.10) and (3.12) depends on the square of the HO_2 radical concentration.

In NO-rich polluted atmospheres, however, reaction (3.6), the rate of which is proportional to the first power of the HO_2 concentration, competes favorably for HO_2 radicals. Reaction (3.6) is very fast in NO-rich atmospheres, with the result that the generation of H_2O_2 in reactions (3.10) and (3.12) is suppressed. Although the uptake of the limited H_2O_2 into cloud water and rain will take place efficiently, for these circumstances the amounts of H_2O_2 may be significantly less than those of HSO_3^- in the water. Obviously, the oxidation of only a fraction of the HSO_3^- can occur for these conditions, and the reaction becomes oxidant limited. SO_2 in cloud water cannot be oxidized faster than the oxidant is provided to the droplet.

Note that for the case of an oxidant-limited reaction, a nonlinear response in sulfuric acid deposition will result from emission reductions. Only when SO_2 emissions are reduced so that ambient concentrations of SO_2 approach the level of the oxidant present in cloud water will a decrease in the sulfuric acid formation and deposition result. For example, if the H_2O_2 available in cloud water were consistently only 40 percent of the SO_2 that is dissolved in the cloud water at a given location, and if oxidation occurred largely through the H_2O_2-HSO_3^- reaction, then a 60 percent reduction of the SO_2 would result in no reduction in the sulfuric acid in cloud water at this location, but subsequent reductions would lead to proportionally lower acid formation and deposition. It is also possible that even with sufficient oxidant in cloud water, other substances that may also be present, such as formaldehyde, may inhibit the H_2O_2-HSO_3^- reaction. Existing analytical data for H_2O_2 in clouds do not allow an unambiguous conclusion to be reached today on the possible importance of these nonlinear effects.

Limitation of oxidant for HSO_3^- or SO_2 may arise because of physical processes as well as the chemical influence described. For example, we have noted previously that it is likely that H_2O_2 vapor present in dry air dissolves in cloud droplets to provide oxidant for the conversion of SO_2. Hydrogen peroxide is a very soluble gas and may be rained out early in some storm systems, leaving a significant fraction of SO_2 vapor unreacted.

Several types of nonlinear effects may be expected from factors not immediately related to oxidant levels. For example, as described in Chapter 2 and Appendix A, there is some evidence that SO_2 is oxidized more readily in the aqueous phase than in the gas phase. Also, increased concentrations of alkaline soil dust in the air due to drought or changing wind patterns can result in the neutralization of precipitation acidity.

In the absence of extensive measurements, we judge that nonlinear effects of SO_2 emission control on acid deposition that arise from chemical conversion mechanisms are probably small for the gas-phase conversion steps, but significant nonlinearity is anticipated for certain special conditions such as an oxidant-limited H_2O_2-HSO_3^- reaction in cloud water. However, these conditions cannot be tested from the existing data base.

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FINDINGS AND CONCLUSIONS

Application of current air-quality models to regional-scale processes has provided guidance on the significance of dynamic processes influencing sulfur deposition. Theoretical models have provided results that are qualitatively consistent with empirical observations, thus demonstrating important temporal and spatial scales of source-receptor relationships. Qualitatively the models have pointed to the importance of certain geographical groupings of SO_2 sources and the potential influence of the sources on certain receptor areas. However, current models have not provided results that give confidence in their ability to translate SO_2 emissions from specific sources or localized groupings of sources to specific sensitive receptors. Little has been done in models to translate NO_x emissions into nitrate deposition or to link sulfate and nitrate to acid (H+) deposition. These capabilitities are considered essential for models to be used to study the consequences of alternative control strategies in circumstances in which long-range transport processes are involved.

Because of the simplifying assumptions that are made in order to develop practical, economical regional-scale models of air quality and because data are not available to validate or verify them, researchers in the field generally have only limited confidence in current results. The models and their results are useful research tools. However, because of deficiencies in the base of meteorological data required as input and because of the sensitivity of their output to simplifying assumptions regarding both the physical and chemical processes, we do not regard currently available models as sufficiently developed to be used with confidence in predicting responses of the atmospheric system to alternative control strategies.

Despite these limitations, theoretical models are and probably will continue to be used in industrial and urban planning, for which spatial scales are smaller than those of interest in acid deposition. Given the state of knowledge of the physics and chemistry of the atmosphere in the context of long-range transport of air pollution, and given the state of the art of techniques for making quantitative estimates, we advise caution in projecting changes in deposition patterns that result from changes in emissions of precursor gases.

On the basis of laboratory evidence, we conclude that an alternative to the model of Rodhe et al. (1981), which has been widely used to represent the chemical processes involved in acid deposition, more correctly employs gas-phase reactions leading to oxidation of SO_2 that results in HO-HO_2-radical chain propagation. Laboratory evidence suggests that chain-terminating reactions involving SO_2 probably play only a minor role in atmospheres polluted with SO_2. When the Rodhe-Crutzen-Vanderpol model is modified so that SO_2 oxidation does not terminate chains, the nonlinearity in the relationship between changes in ambient SO_2 concentrations and changes in ambient sulfate concentrations (i.e., the commonly reported result of the Rodhe model) is greatly reduced.

Laboratory and field studies as well as theory suggest that oxidation of SO_2 in cloud water is rapid and complete, provided that concentrations of oxidants (H_2O_2, O_3) are sufficient (see Chapter 2). Measurements of oxidant concentrations in cloud water, although limited, suggest that concentrations may be sufficient in eastern North America for complete oxidation of SO_2, except perhaps in winter. If this is the case, then strong deviations from linearity in the relationship between changes in annual average ambient SO_2 concentrations and changes in the net production of sulfate in clouds would not be expected.

The relationships between emissions of SO_2 and NO_x and the deposition of sulfuric and nitric acids are complex. Models to predict patterns of the deposition of hydrogen ion will have to account for neutralizing substances as well as sulfuric and nitric acids. Assuming that the ambient molar concentrations of NO_x and basic substances (such as ammonia and calcium carbonate) remain unchanged, we conclude that a reduction in sulfate deposition will result in at least as great a reduction in the deposition of hydrogen ion.

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4 Empirical Observations and Source-Receptor Relationships

The analysis of data taken in the field complements the development of theoretical models as a means for understanding both the phenomenon of acid deposition and the responses of the atmospheric system to alternative emission-control strategies. Field measurements not only reveal insights into the nature of the atmospheric processes involved in the deposition of acid-forming materials but also may hold the greater promise of providing direct, unequivocal evidence from which responses to mitigating strategies might be assessed. Data from which the spatial and temporal distributions of SO_2 and NO_x over large regions of North America might be derived have not been available until recently. Similarly, reliable sampling of the chemistry of precipitation in North America is a recent development. Even so, sampling of ambient pollutants in the atmosphere and sampling of those in precipitation generally have not been simultaneous. There are no direct measurements of regional dry deposition for gases or particles. [For a description of the monitoring systems in North America and data interpretation, see U.S./Canada Work Group #2 (1982).]

Although data are relatively sparse in North America, those that are available tell us much about the phenomenon. A more extensive data base exists for northern Europe, where atmospheric SO_2 and sulfate in precipitation have been monitored systematically for several decades. Much of our understanding of acid deposition has come from analysis of the European experience and the transfer to and replication of those data on this continent. That being so, differences in patterns of emissions, climatic factors, ground cover, and influences of marine air between Europe and North

America require that our understanding be tested against North American data.

In this chapter we review the existing data for North America and use the data to assess the extent to which the relationships between emissions and deposition can be inferred from observations. The limitations of available data, based on conventional sampling and measurement methods, are discussed first. Results from some pertinent field programs are surveyed to illustrate the importance of meteorological processes for the variability of ambient air pollutants and acid-forming components in precipitation. Results of statistical analyses of data on ambient concentrations of pollutants are reviewed, and data on both emissions and deposition are analyzed to infer the influence of sources on deposition.

Our survey is not a comprehensive one; it focuses on the results that bear most directly on atmospheric transport and transformation. In this context, the issues addressed are whether data are sufficient to infer (1) the extent to which a given region of sources affects receptor sites in remote locations and (2) the importance of nonlinear processes in determining the relationships between the magnitudes of emissions in source regions and the quantities of acid-forming substances deposited in receiving regions.

One of the greatest difficulties in establishing relationships between sources of pollution and conditions at receptors is accounting for the influences of atmospheric processes on the behavior of pollutants. The atmospheric processes involved include airflow, mixing, and chemical transformations. These processes are responsible, directly or indirectly, for the distribution and rate of deposition. Attempts to discern the influences of atmospheric processes on source-receptor relationships have taken different routes, including (1) descriptive accounts of observations, (2) analysis of data segregated by airflow from source areas (trajectories), (3) statistical analyses of data, and (4) inference from source tracers. In this chapter we describe the approaches used in these types of analyses and the results obtained. We also analyze existing data on emissions and deposition to discern trends and the relative behavior of sulfur and nitrogen emissions in the atmosphere.

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AEROMETRIC DATA AND THEIR LIMITATIONS

Most of the historical data on ambient air concentrations describe urban conditions. A large body of monitoring data exists in the National Aerometric Data Base (NADB), but few of these observations have been analyzed or interpreted. One of the more reliable and complete data sets available that describes regional air quality in the eastern United States was taken during a single year, 1977-1978: the Sulfate Regional Experiment (SURE) (Mueller and Hidy 1983). Unfortunately, few precipitation chemistry data were collected during the period of the study. In contrast to other data sets, those obtained from the SURE experiment have been analyzed in detail. Complementary data are or will be available from the western United States for 1980-1982 as a result of several studies (Allard et al. 1981, Pitchford et al. 1981). Observations of precipitation chemistry have been made periodically in the mid-1950s, from 1959 to 1966, and from 1972 to date in the programs described, for example, by Wisniewski and Kinsman (1982). Air-quality and precipitation data have been collected in Canada since 1974 in the Canadian (CANSAP) monitoring program.

Precipitation data provide a direct measure of wet deposition. Since there are no direct measurements of dry deposition, regional patterns are estimated from ambient concentrations and deposition velocities (Appendix C). Since deposition velocities depend on the airflow near the surface, surface properties, and cover, these calculations are believed to be uncertain for quantitative evaluation.

The quality of the data describing regional-scale processes is variable. The precision and accuracy of the measurements are generally not well defined in work reported prior to the late 1970s. Recent programs, however, have made considerable progress in reporting data with supplemental information on the calibration of instruments, as well as the errors contained in the observations. A definitive discussion of aerometric data is included in Mueller and Hidy (1983). The quality of recent data on wet deposition is less formally documented, but extensive information on the comparability of data is available from the Illinois Water Survey Laboratories, the Department of Energy's Environmental Measurement Laboratories in New York, and the Environmental Protection Agency's Environmental Monitoring Systems Laboratory. A statistical analysis of data from two independent

precipitation sampling networks (SURE and MAP3S) covering a similar region in the eastern United States indicated that there is good agreement between data on the concentrations of the major ions, H^+, sulfate, nitrate, and ammonium, between the two networks (Pack 1980).

Documentation of the quality of data is essential to ensure that comparisons will be possible with data from future studies. Experiments and monitoring programs should incorporate such efforts to avoid or at least minimize controversies about data interpretation. The integrity of the sampling and analytical methods employed is another important consideration in measurement quality. Despite years of effort in the development of methods, the instrumentation used in past and current programs is subject to serious interferences. Uncertainties in results can be large. Interferences or ambiguities in the methods are particularly serious under rural or remote conditions, in which concentrations of pollutants or deposition levels are low.

Listed in Table 4.1 are examples of sampling or measurement uncertainties that exist in currently available methods of sampling and analysis. From the table it is evident that few of the important chemically related measurements can be made without ambiguity because of uncertainties in the methods.

RELATIONSHIPS AMONG AEROMETRIC PARAMETERS

Atmospheric measurements provide a basic description of spatial and temporal distributions of pollutants. On a regional scale, the distribution of SO_2 shows strong gradients near sources, but airborne sulfate shows relatively weak gradients. Concentrations of both SO_2 and $SO_4^=$ are elevated over the industrialized or urbanized parts of the eastern United States (Figures 4.1 and 4.2). High concentrations of sulfur oxides are found through the Ohio River Valley into Pennsylvania and New York to the Atlantic Coast. From this region of high concentration, there is a strong gradient in ambient SO_2 concentration northeastward toward the Adirondacks, Vermont, and New Hampshire. The distribution of nitrogen oxide concentrations is generally similar to that of sulfur oxides, but it may show more localized maxima near urban areas (e.g., Mueller and Hidy 1983).

Paralleling the distributions of ambient concentrations are the distributions of sulfate and nitrate in precipi-

TABLE 4.1 Uncertainties in Conventional Sampling and Analytical Methods

Parameter	Method	Comments
Sulfur dioxide	Flame photometric	Water vapor and CO_2 interference near detection limit
Particulate sulfate	Filter substance collection and wet extraction and ion chromatography	Potential SO_2 adsorption on sample or filter substrate, especially glass-fiber material
Nitrogen dioxide	Chemiluminescent	Interference from HNO_3, N_2O_5, organic nitrates
Nitric acid	Nylon absorber/wet extraction; denuder tubes	NH_4NO_3 volatility; possible interferences
Nitrogen pentoxide	Method not available	—
Particulate nitrate	Filter collection/wet extraction and ion chromatography	Potential N_xO_y and HNO_3 vapor adsorption on filter mat, particularly glass fiber
Ammonia	Absorption on impregnated filter/water extraction and ion chromatography	Uncertain absorption efficiency
Acidity in aerosol	NH_3 denuder tubes with filter collection/wet extraction and acid-base titration	Potential interferences from ambient ammonia absorption from basic particles
Wet deposition rate	Wet collector/wash and chemical analysis	Seal on bucket can leak contaminating sample, collector may fail to open on small rainfall
Sulfate in water	Ion chromatography	Reliable technique
Nitrate in water	Ion chromatography	Reliable technique
Acidity in water	pH meter	Care must be taken on multiple, repeated electrode calibration and wash
Dry deposition of gases	Chemical absorption plate	Geometry dependent, does not simulate vegetation
Dry deposition of particles	Flux meter	Insufficient time resolution to be compatible with turbulent fluctuations

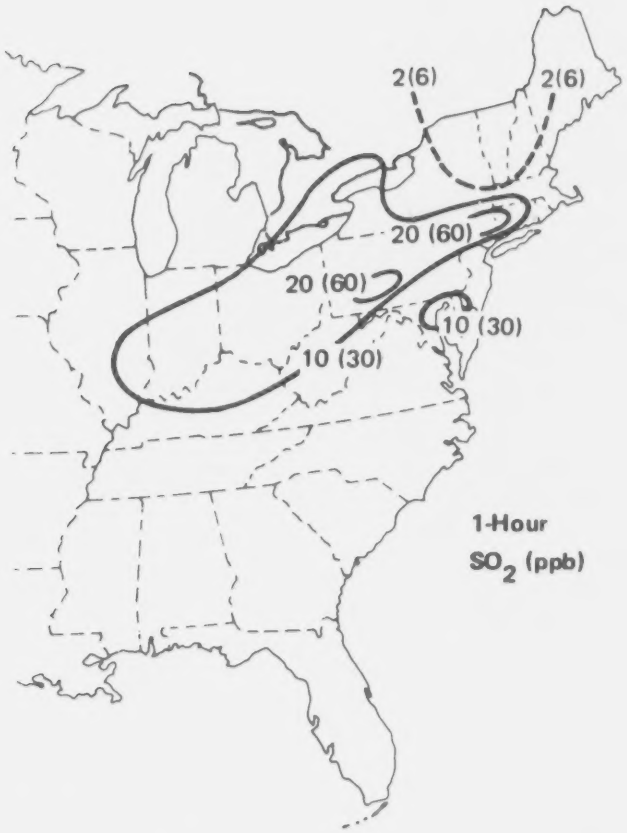

FIGURE 4.1 Composite spatial distribution of 1-hour average concentrations (ppb) of SO_2 from one representative month in each season between 1977 and 1978. This average is approximately equivalent to an annual average. Dashed isopleth is based on limited quantities of SURE data for 1977-1978 and on data taken at Whiteface Mountain, New York, after 1978. Numbers in parentheses are calculated values of dry deposition rates in kilograms of sulfur per hectare per year assuming a uniform deposition velocity of 0.8 cm/s. SOURCE: Data on concentrations from Mueller et al. (1980) and, for Whiteface Mountain, from V. Mohnen, State University of New York, Albany, personal communication (1983).

tation, as indicated in Figures 4.3 and 4.4. These distributions may be compared with that of the hydrogen ion concentration in precipitation in Figure 4.5. From these figures, the pattern of deposition of hydrogen ion in precipitation appears to correspond to regions of elevated sulfate and nitrate concentrations. This finding does not necessarily follow without accounting for all the cations (such as NH_4^+ and Ca^{++}) and anions that

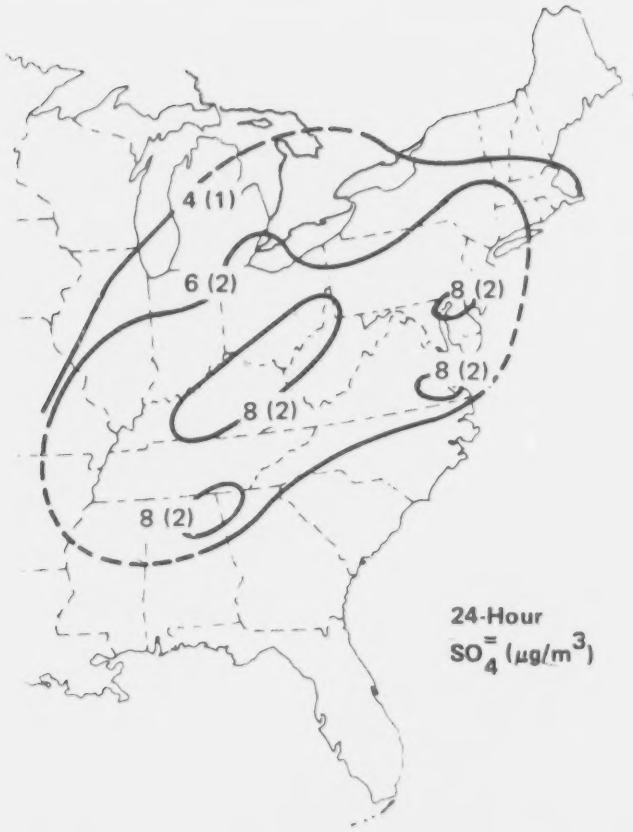

FIGURE 4.2 Composite spatial distribution of 24-hour average concentrations of sulfate ($\mu g/m^3$) from one representative month in each season between 1977 and 1978. This average is approximately equivalent to an annual average. Numbers in parentheses are calculated values of dry-deposition rates in kilograms of sulfur per hectare per year assuming a uniform deposition velocity of 0.2 cm/s. SOURCE: Data on concentrations from Mueller et al. (1980).

may be important factors affecting the acidity of precipitation.

A qualitative comparison between dry- and wet-deposition rates can be made from observation. Wet-deposition rates for sulfate are shown in Figure 4.3 for data taken in 1980. Estimated dry-deposition rates for SO_2 and particulates are shown in parentheses in Figure 4.1 and 4.2 (estimated values in kilograms of sulfur per hectare per year). The rates have been calculated using uniform deposition velocities considered typical of values reported in the literature for SO_2

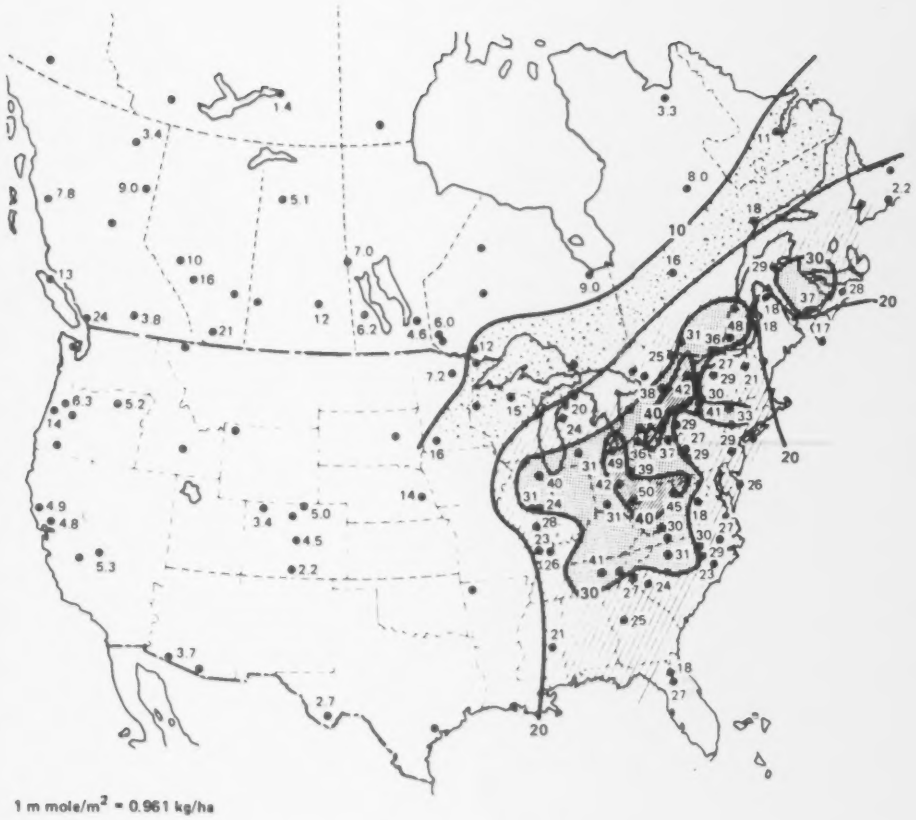

FIGURE 4.3 Spatial distribution of mean annual wet deposition of sulfate weighted by the amount of precipitation in North America in 1980 (mmoles/m^2). SOURCE: U.S./Canada Work Group #2 (1982).

gas and for submicrometer particles (sulfate). Note that in much of the region of high ambient concentrations, dry deposition apparently dominates total deposition. This result suggests that dry deposition of SO_2 exceeds wet deposition in parts of the Ohio River Valley and the Ohio-Pennyslvania-western New York area but becomes progressively less important farther from the region of major emissions, to the northeast in New England and Canada. At large distances from sources, ambient concentrations of sulfur oxides are low; wet deposition will dominate dry deposition far from sources if precipitation is significant.

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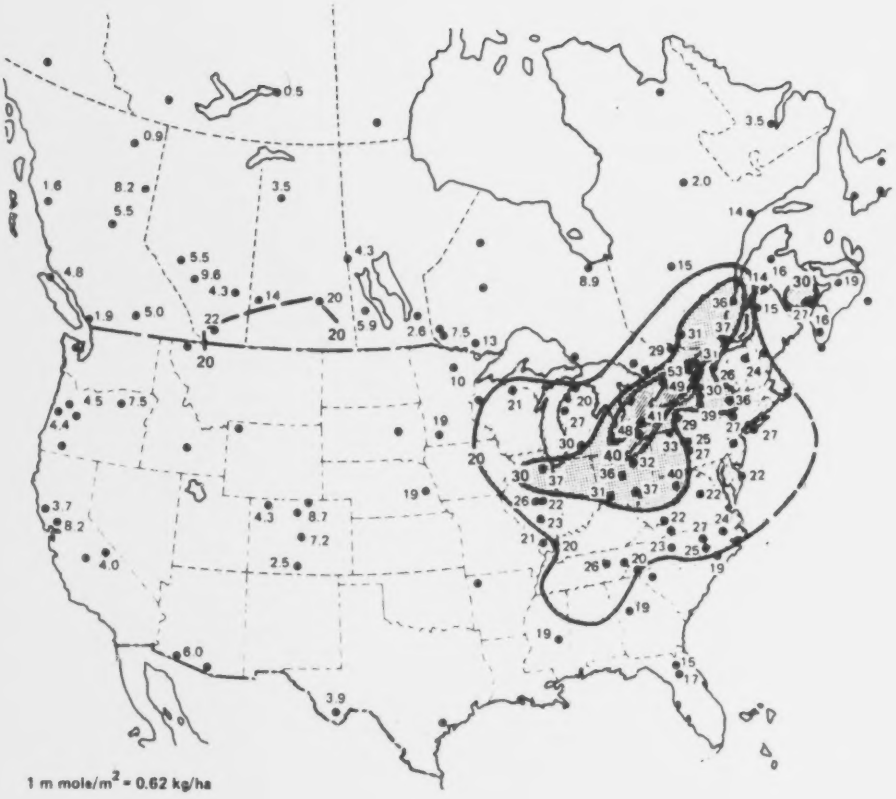

FIGURE 4.4 Spatial distribution of mean annual wet deposition of nitrate weighted by the amount of precipitation in North America in 1980 (mmoles/m^2). SOURCE: U.S./Canada Work Group #2 (1982).

THE INFLUENCE OF METEOROLOGICAL CONDITIONS

The spatial distributions of deposition for acid-forming materials are similar, and elevated concentrations appear to be associated with areas in which emissions are high. The temporal behavior of sulfur oxides and nitrogen oxides is dominated by meteorological variability. For example, analysis of the SURE data indicates that the variability in ambient concentrations of SO_2 was as much as an order of magnitude greater than the variability in SO_2 emissions over the eastern United States (Mueller and Hidy 1983). This finding suggests that advanced and sophisticated analyses are needed to separate the influences of emissions from those of aerometric parameters in such data. These analyses have

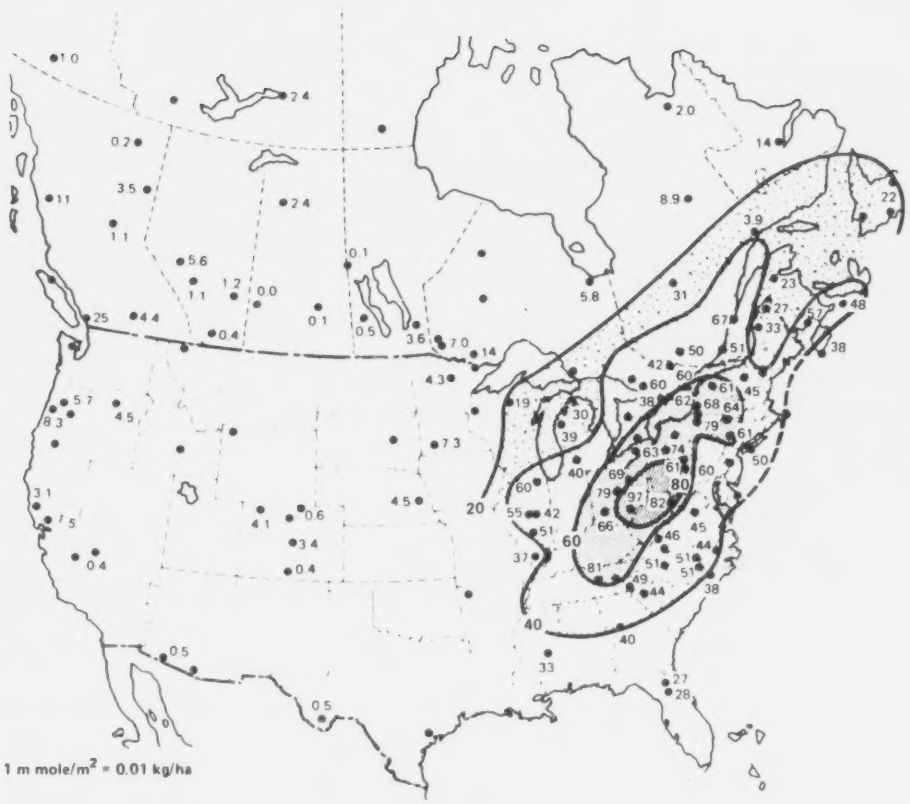

FIGURE 4.5 Spatial distribution of mean annual wet deposition of hydrogen ion weighted by the amount of precipitation in North America in 1980 (mmoles/m^2). SOURCE: U.S./Canada Work Group #2 (1982).

followed two directions in the recent literature. The first is descriptive, taking into account the behavior of sulfur oxides in the atmosphere as a function of climatological or meteorological features. The second stems from the first by adapting statistical techniques to separate influences of different processes. In this section we review results of analyses of data according to meteorological conditions and studies of air-mass trajectories. In the next section we present the results of statistical methods of analysis.

Classification of Meteorological Conditions

One of the primary conclusions of many studies of field observations is the absence of a definitive relationship

between SO_2 emissions and sulfate concentrations in dry air or in precipitation patterns. Sulfur emission rates are relatively constant, whereas the concentrations of sulfate aerosol and SO_2 in the air are highly variable and dominated by meteorological conditions (Electrical Power Research Institute 1981). The concentrations of sulfate particles tend to be high in summer, presumably because of more rapid photochemical oxidation of SO_2, high in the central and western region of high-pressure systems that move slowly toward the Atlantic, and high in maritime tropical air emanating from the coastal region of the Gulf of Mexico. Concentrations of SO_2 also tend to be high under stagnant air conditions of slow-moving, high-pressure, anticyclonic systems but tend to be low in the maritime tropical air. Trajectory analyses applied to specific transient anticyclonic systems have further demonstrated the tendency of such systems to favor air-stagnation situations in which high levels of pollutants accumulate (King and Vukovich 1982).

Evidence based on chemical and meteorological analyses as well as satellite photos suggests that, on occasion, pollutants originating in the Midwest and Northeast can be entrained in the clockwise flow around a transient high-pressure system and transported to the Gulf Coast region. The pollutants are believed then to be advected back through the Midwest and Northeast in the southwesterly flow of maritime tropical air (Wolff et al. 1981, 1982). The slow-moving flow of southwesterly maritime air from the southern states entrains pollutants from sources in its path and eventually moves over the northeastern United States and eastern Canada. Summertime convective storms that occur in this air tend to be quite acidic compared with precipitation during the cooler months (MAP3S/RAINE Research Community 1982, Raynor and Hayes 1982a).

Precipitation during autumn, winter, and spring in eastern North America also tends to occur in moist southwesterly air that frequently emanates from the southern states. During these seasons, most of the precipitation occurs in the vicinity of warm fronts associated with cyclonic low-pressure storm systems. Precipitation develops because the southwesterly air travels faster than the warm front, which represents the boundary of colder and heavier air lying to the north of it. The southerly air ascends over the cold air and is cooled, producing large areas of precipitation. The situation is illustrated in Figure 4.6. The center of

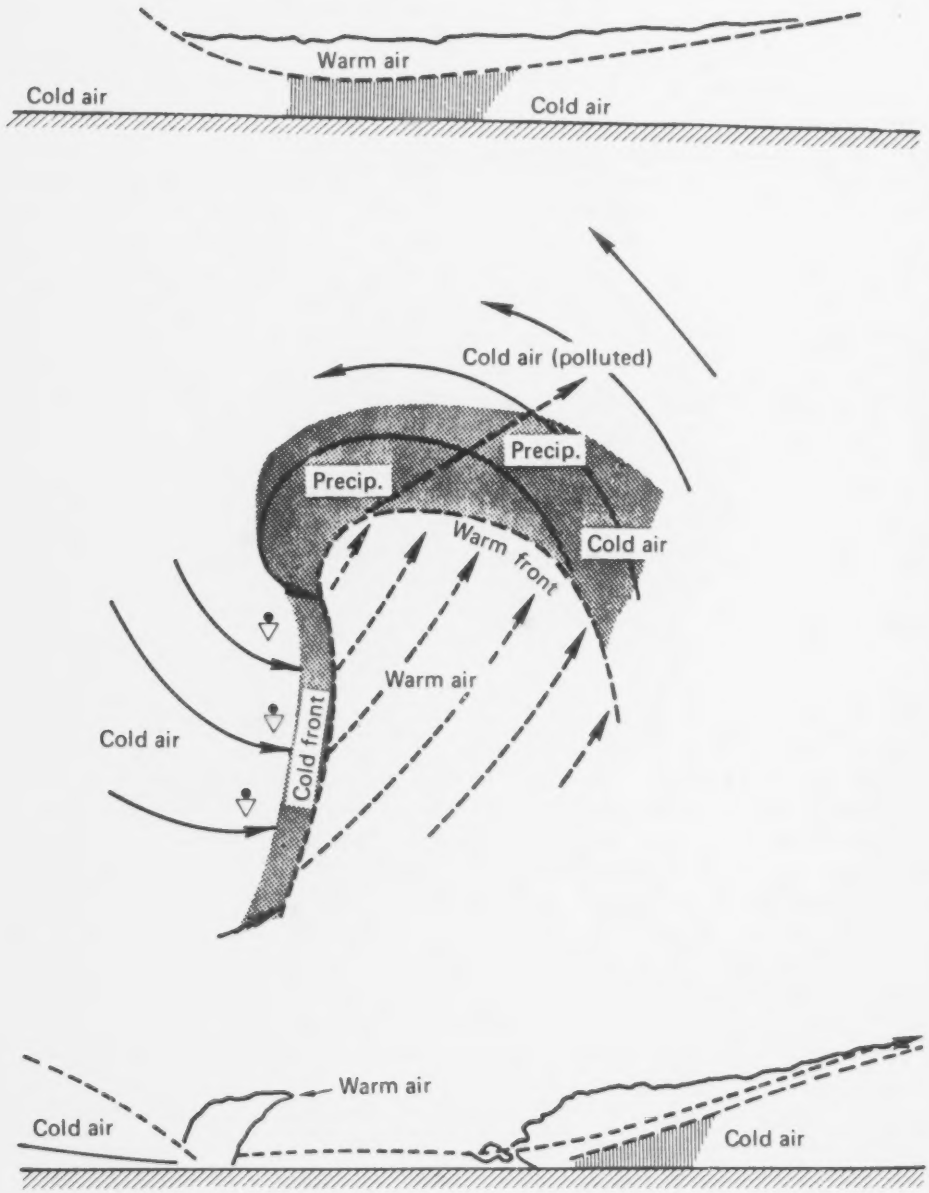

FIGURE 4.6 Idealized depiction of air masses and precipitation typically associated with warm fronts and low-pressure systems during autumn, winter, and spring in eastern North America. SOURCE: Lazrus et al. (1982).

the low-pressure system tends to travel toward the Northeast, and naturally the associated warm front moves with it. A map of preferred tracks of cyclonic weather systems (Figure 4.7) indicates the tendency for movement toward the Northeast. The bands of precipitation associated with the warm front move northeastward, depositing pollutants incorporated by the southwesterly air during its passage from southern or midwestern states. Although warm frontal precipitation is less acidic than summertime convective rain, warm fronts may deposit more acidic material in the Northeast because they deposit more precipitation (Raynor and Hayes 1982a). Figure 4.8 shows precipitation and wet deposition of chemical species as a function of the type of precipitation and type of synoptic weather system at Upton, New York.

The data of the SURE network have made it possible to quantify pollution episodes in terms of duration, sulfate concentration, extent of area, frequency of occurrence, and association with meteorological conditions. This information is summarized in Tables 4.2 and 4.3. Table 4.2 defines events (episodes) according to the number of stations with sulfate readings in specific ranges. The notations in Table 4.3 referring to meteorological type are illustrated in Figure 4.9. Warm southwesterly maritime tropical air (indicated as mT) referred to above occurs about 12 percent of the days in the region of the SURE network. The air involved in warm frontal precipitation is included in a transitional category indicated as Tr in Figure 4.9 and Table 4.3. Air masses of continental polar origin (cP) are identified with large areas of high barometric pressure and anticyclonic circulation. Continental polar cold (cPk) air refers to flow from central Canada southwest. Air-mass stagnation under the high pressure area is referred to as cP2 conditions, while cPw refers to warm airflow from south to north on the west side of the anticyclone.

Recent studies of fluctuations in SO_2 and sulfate concentrations along with other evidence indicate that sulfate aerosol advected from distant origins makes a large contribution to the particulates in the New York City area only during the summer (Lioy and Morandi 1982, Tanner and Leaderer 1982).

A correlation analysis of sulfate particle concentrations in dry air between various SURE sites indicates that significant regions of correlation generally extend 200 to 300 km from major SO_2 sources during pollution episodes and rarely beyond 500 km (Electric Power Research

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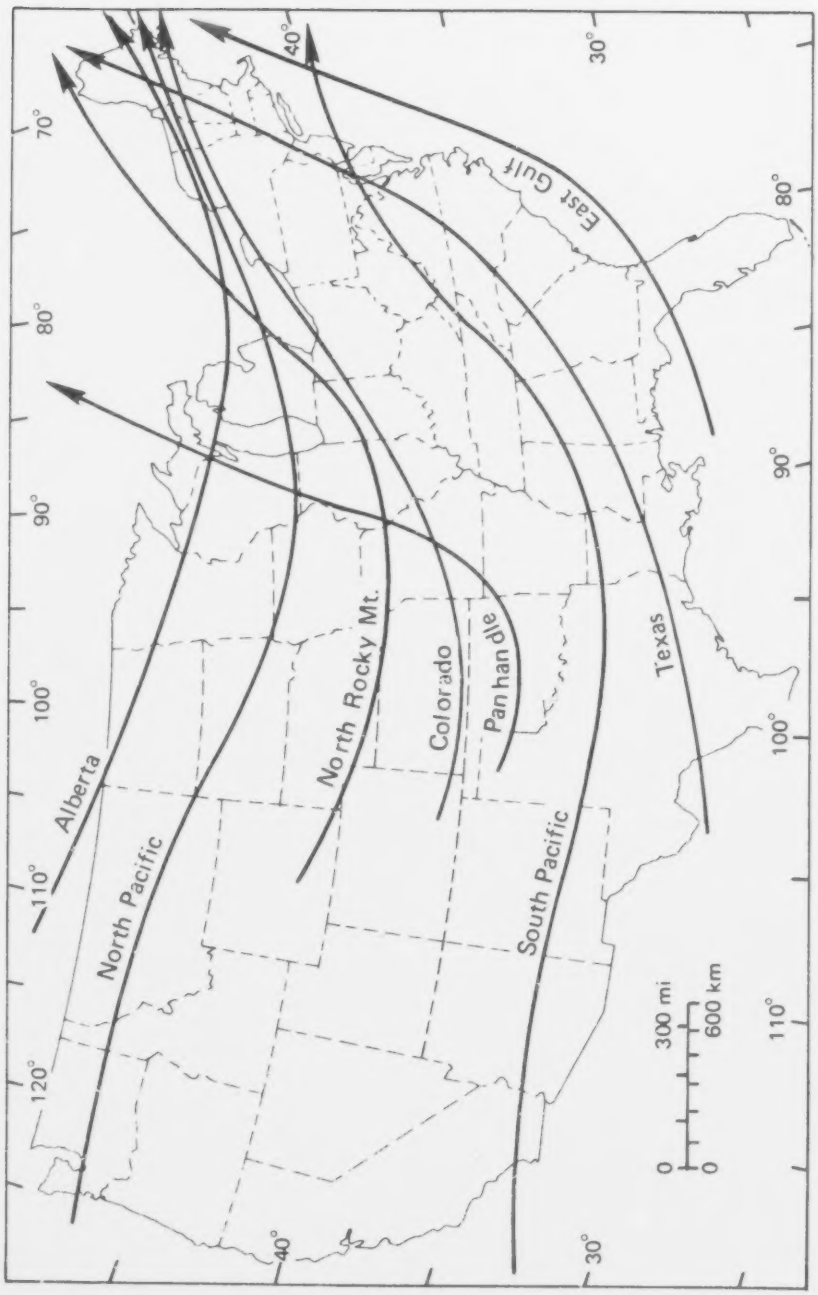

FIGURE 4.7 Main tracks of cyclonic (low-pressure) systems across the United States. SOURCE: Eichenlaub (1979).

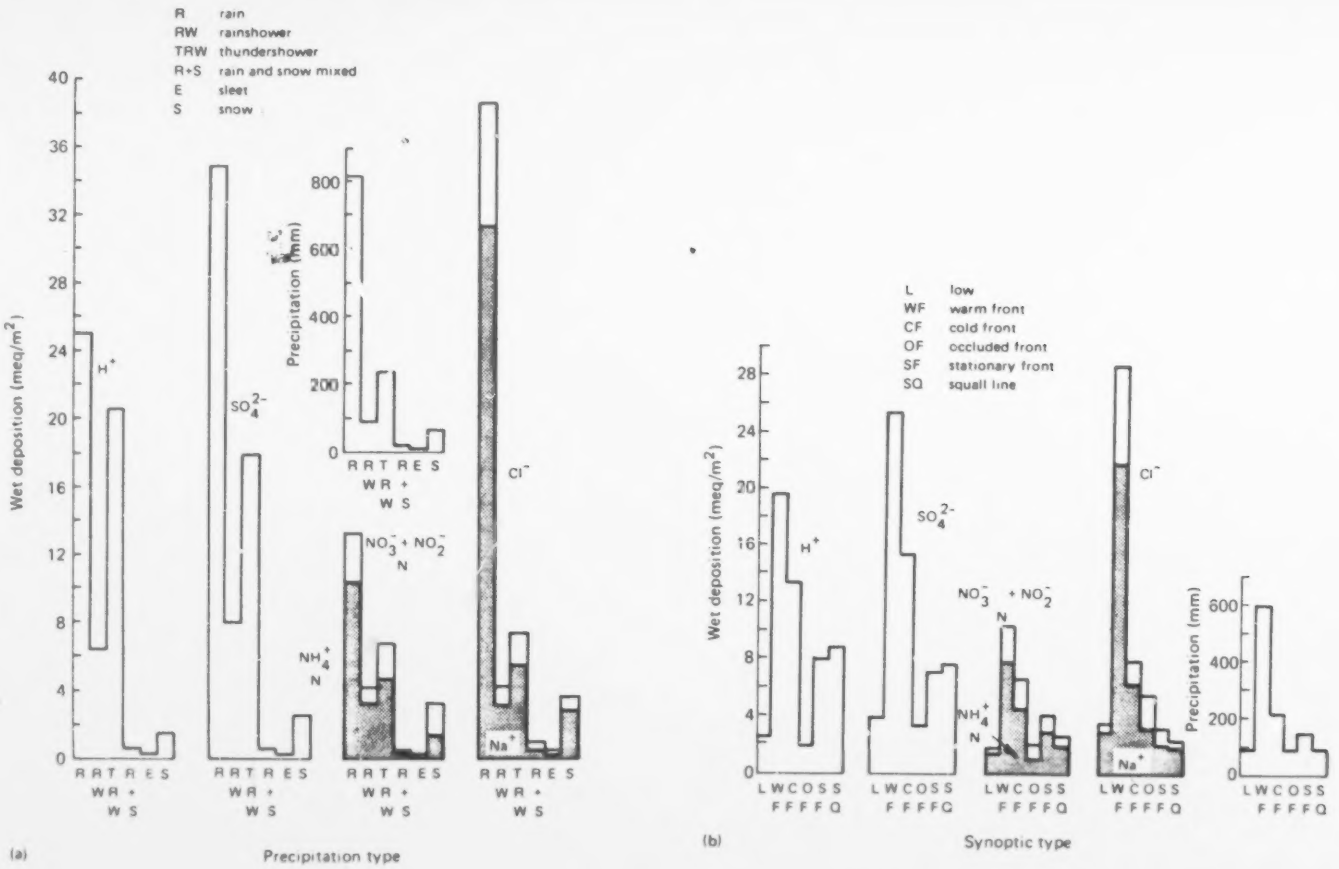

FIGURE 4.8 Precipitation and wet deposition of chemical species as a function of (a) precipitation type and (b) type of synoptic weather system at Upton, New York. SOURCE: Raynor and Hayes (1982a).

TABLE 4.2 Definitions of Pollution Events by Intensity and Coverage of Sulfate Concentration

Event Group	Percentage of Stations with Sulfate Concentrations			Days per Event—	
	>10 mg/m³	>15 mg/m³	>20 mg/m³	Range	Mean
Enlarged regional	40 to 93	25 to 72	10 to 61	3 to 11	5
Regional	40 to 70	15 to 25	5 to 10		
Subregional	25 to 50	5 to 15	<5	1 to 7	2
Nonregional	<35	<5	0	—	—
No event	0	0	0	—	—

SOURCE: Electric Power Research Institute (1981).

Institute 1981). The exception to this observation occurs during ducting of pollutants from southern regions to the Northeast by southwesterly air.

Another indication of the relative importance of meteorological processes in determining wet deposition can be found in European data. In eastern North America, the warmer months tend to be the period of greatest precipitation as well as of most rapid photochemistry. At certain coastal sites in Europe, however, the season of greatest precipitation does not correspond to the season of most rapid photochemistry. Figure 4.10 shows the seasonal variations in sulfate (in excess of that from sea salt) and amount of precipitation for three groups of sites in Europe. The triangles indicate coastal sites where periods of greatest precipitation do not correspond to the period of greatest sunlight intensity and hence to accelerated photochemistry. These cases demonstrate that gas-phase photochemistry may not be the dominating factor controlling the seasonal trend of wet deposition of sulfate. The influence of meteorological and precipitation processes (and possibly of aqueous chemistry) appears to be of major importance.

Air-Mass Trajectories

The influences of various source regions on given receptor sites have been calculated by following the trajectories of the air parcels bearing the pollutants. The most widely used method calculates the trajectories along isobaric surfaces. The inaccuracies of the method arise mainly from the assumption that vertical air motions can be ignored, because the necessary meteorological data on which the calculation depends are available only twice

TABLE 4.3 Annual Percentage of Event Days by Air-Mass Category

Event Group	Air-Mass Category[a] cPk	cP2	cPw	Tr	mT	Annual Percentage of Days in Each Event Group
Regional	0	12	7	2	9	30
Subregional	0	4	4	4	3	15
Nonregional	2	10	2	18	0.5	32.5
No event	6	4	0.5	12	0	22.5
Annual percentage of days in each air mass	8	30	13.5	36	12.5	100

SOURCE: Electric Power Research Institute (1981).
[a]See Figure 4.9 and the text for descriptions of the air-mass categories.

daily at widely spaced locations, and from the difficulties associated with calculating trajectories in the vicinity of precipitating systems. The periods over which samples of precipitation are collected are usually so long (one week or one month) that the source regions of the air masses contributing to the sample may change during a single collection period. This problem may even arise in event sampling in the case of cyclonic storm systems in which the precipitation falls from air masses that frequently change markedly over a period of hours.

Recent studies of air-mass trajectories at Whiteface Mountain in the Adirondacks (Wilson et al. 1982) indicate that 62 percent of the sulfate ion and 65 percent of the nitrate ion are deposited by precipitation associated with air parcels emanating from the Ohio River Valley and midwestern states (Figure 4.11). Wilson et al. also point out that 56 percent of the total annual precipitation at Whiteface Mountain is carried by those air parcels and that the wet deposition of acid may be related more to this fact than to the higher concentrations in the precipitation originating upwind to the south and southwest. They also point out that the geographical gradient of acid deposition (normalized to the amount of rainfall) is not very steep. The normalized depositions vary from 35 mg/m^2 of sulfate at the source region to 27 mg/m^2 at Ithaca, New York, and University Park, Pennsylvania, to 18 mg/m^2 at Whiteface Mountain. They postulate that atmospheric mixing of pollutants together with varying patterns of precipitation tend to make the spatial distributions of pollutant material deposited per unit amount of precipitation relatively uniform when considered over time scales of a year or more.

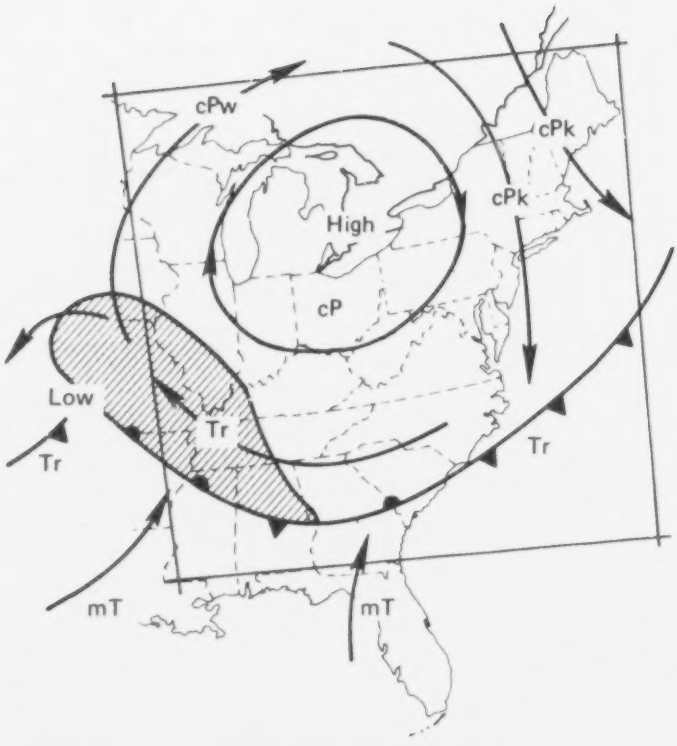

FIGURE 4.9 Categories of air masses used to classify regional air quality in Table 4.3. SOURCE: Electric Power Research Institute (1981).

The tendency of atmospheric mixing processes and varying precipitation patterns to attenuate the gradients expected between source and receptor regions is illustrated by isopleth maps of annual deposition of hydrogen ion (Figure 4.5), sulfate (Figure 4.3), and nitrate (Figure 4.4), which place most of the Adirondacks, a receptor region, in the same isopleth area as the Ohio River Valley, a source region.

A similar trajectory study at Ithaca, New York (Henderson and Weingartner 1982), yielded the percentage deposition for trace constituents in precipitation as a function of the trajectory of the air parcel and time of year. From October through March the percentage depositions from the southwest quadrant were H^+, 72 percent; $SO_4^=$, 77 percent; NO_3^-, 70 percent; and NH_4^+, 84 percent. Of the total precipitation during the period, 71 percent emanated from the southwest quadrant. Similarly, the values from April to September were H^+, 63

percent; $SO_4^=$, 68 percent; NO_3^-, 63 percent; NH_4^+, 76 percent; and precipitation, 52 percent.

It is interesting to note the following: (1) during the cool period the percent deposition of acidic substances corresponds closely to the percent deposition of precipitation; (2) during the warm period southwesterly precipitation deposits a higher percentage of acid per unit percentage of precipitation; (3) the Midwest is an important source of the ammonium ion (derived from gaseous ammonia), which tends to neutralize sulfuric acid; and (4) nitric acid, derived from NO_x emissions that are partially due to vehicular traffic (about 44 percent of NO_x emissions nationwide), is associated with southwesterly precipitation systems. The authors found no direct correlation between emissions and depositions but were able to develop empirical relationships with rainfall rate and air-mass velocity that

FIGURE 4.10 Seasonal variation in deposition of excess sulfate (solid curve) and amount of precipitation (dashed curve) for three groups of stations in Europe. SOURCE: Granat (1978).

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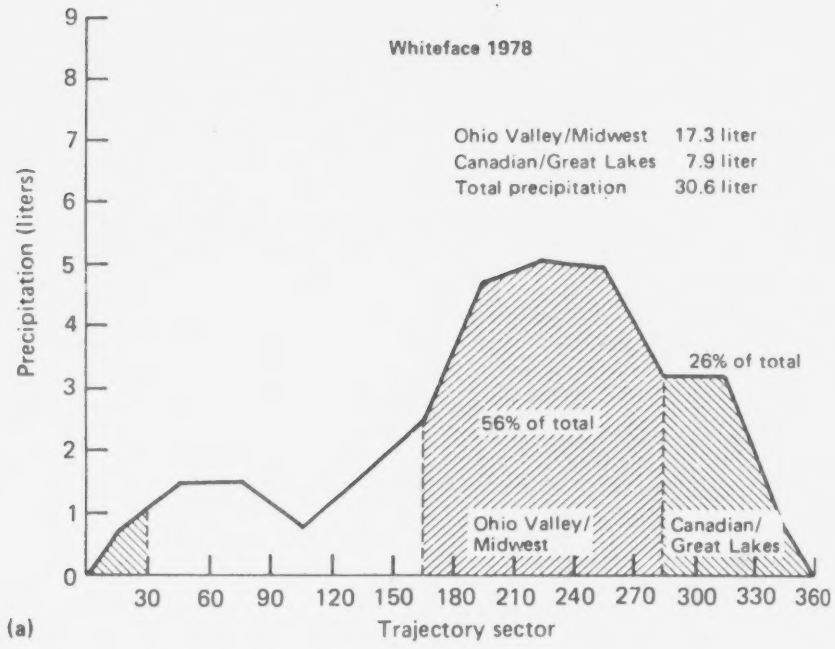
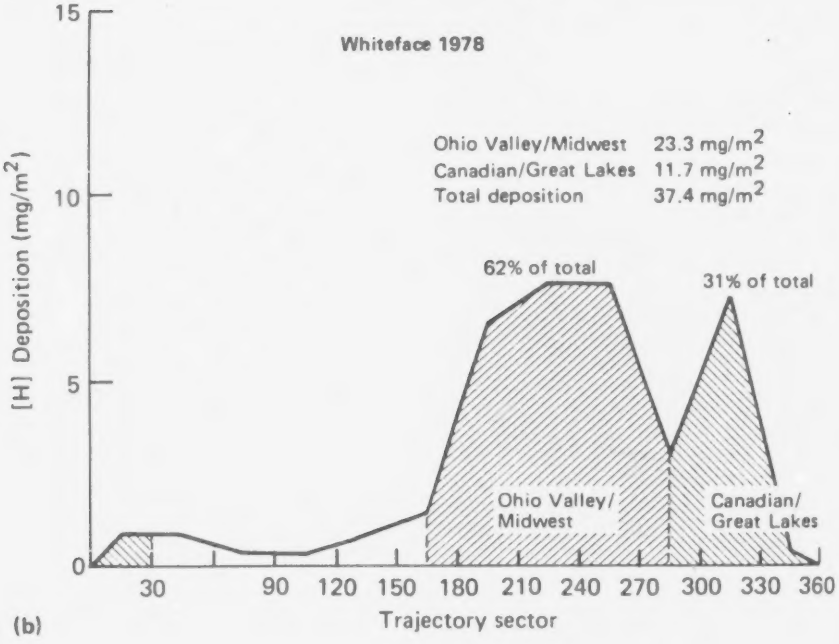
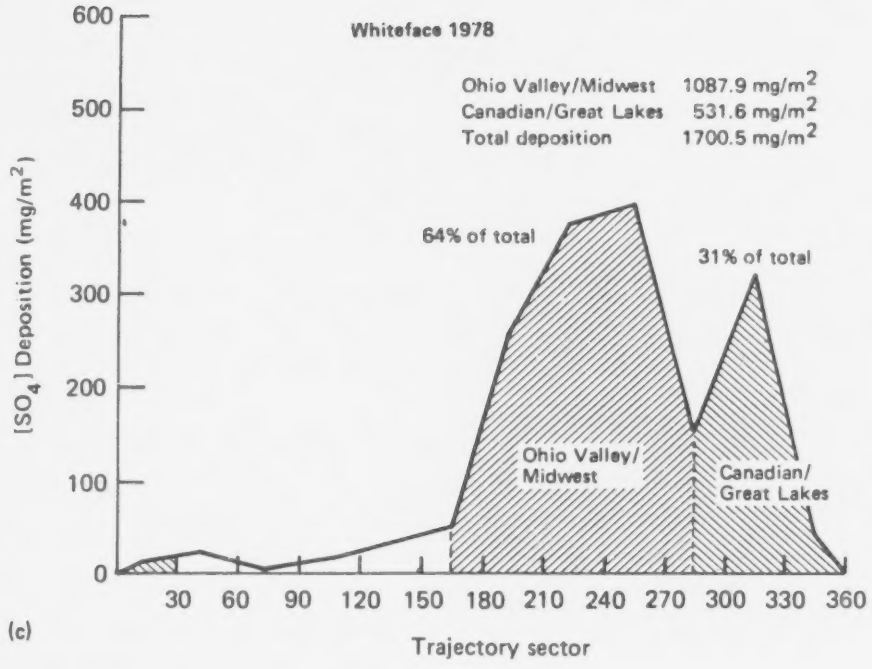

Whiteface 1978
Ohio Valley/Midwest 17.3 liter
Canadian/Great Lakes 7.9 liter
Total precipitation 30.6 liter
Precipitation (liters)
26% of total
56% of total
Ohio Valley/ Midwest
Canadian/ Great Lakes
Trajectory sector
(a)


Whiteface 1978
Ohio Valley/Midwest 23.3 mg/m2
Canadian/Great Lakes 11.7 mg/m2
Total deposition 37.4 mg/m2
[H] Deposition (mg/m2)
62% of total
31% of total
Ohio Valley/ Midwest
Canadian/ Great Lakes
Trajectory sector
(b)



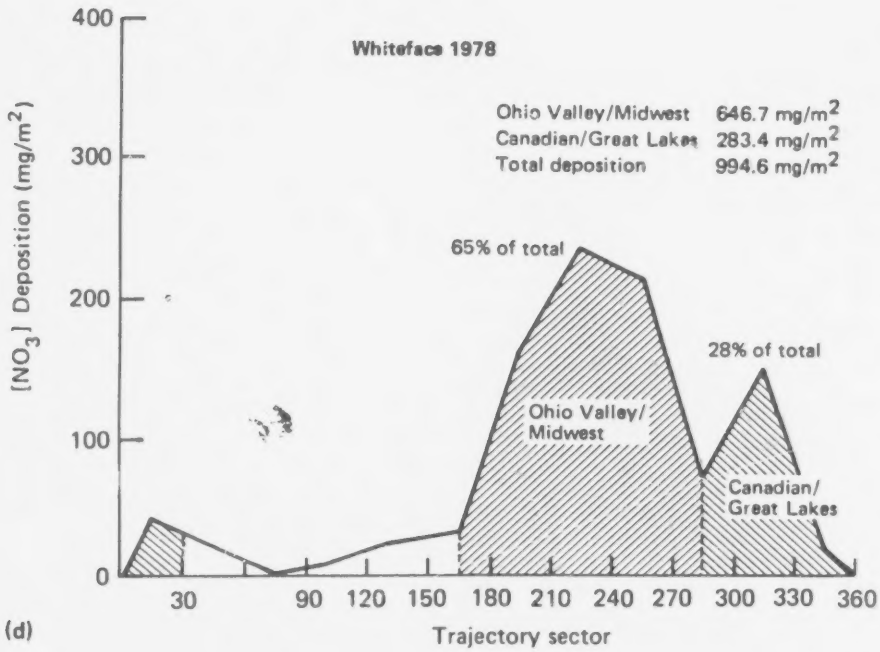


FIGURE 4.11 (a) Precipitation, (b) hydrogen ion concentration, (c) sulfate ion concentration, and (d) nitrate ion concentration in precipitation as a function of the directional sector through which the air parcel passed to reach Whiteface Mountain, New York, in 1978. SOURCE: Wilson et al. (1982).

accounted for 92 percent of the variability of sulfate deposition with SO_2 emissions along the air-parcel trajectory. Similarly, 93 percent of the variability of nitrate deposition with NO_2 emissions was accounted for in these empirical relationships.

A third recent air-parcel trajectory study conducted in south central Ontario showed similar directional expectancies. During the warm period the depositions associated with the southerly and southwesterly air-mass trajectories were H^+, 75 percent; $SO_4^=$, 70 percent; and NO_3^-, 57 percent; and during the cold period, H^+, 85 percent; $SO_4^=$, 87 percent; and NO_3^-, 86 percent (Kurtz and Schneider 1981). The percentage of total precipitation emanating from the southerly and southwestern sectors was not indicated.

The evidence of the air-parcel trajectory studies at specific receptor sites in remote areas of the northeastern United States suggests that the industrialized region to the southwest is a major source of both nitric and sulfuric acids deposited at the sites. However, the high percentage of acid deposition in the Northeast associated with air parcels coming from the southwestern sector may be a result of the high percentage of total precipitation that is delivered by these parcels. Moreover, observed gradients of deposition demonstrate that atmospheric mixing processes and precipitation variability obscure the source-receptor relationship to the extent that simple transport models may not realistically relate specific source regions with specific receptor sites.

STATISTICAL METHODS OF ANALYSIS

Statistical techniques can help to develop additional perspective on the significance of meteorology and chemical processes on the behavior of ambient sulfate. The resulting empirical models are used to relate emissions and ambient conditions.

Conventional statistical techniques have been used to establish correspondences between explanatory variables such as ambient SO_2 or NO_x concentrations, other aerometric variables, and the end products of atmospheric reactions (sulfate or nitrate). In interpreting the data, it is assumed that ambient concentrations of SO_2 or NO_x are proportional to emissions. Linear regression analyses have been performed to relate covariations in the end products to covariations in the explanatory

variables. The results (estimates of regression coefficients) were sometimes inconsistent from location to location and often indicated negative correlations among observables that should in theory be positively correlated. Although misleading estimates may result for a number of reasons, one of the more important problems in aerometric data is collinearity among the explanatory variables. Belsley et al. (1980) discussed this problem, which results when one or more columns of the matrix of the explanatory (or independent) variables is an approximate linear combination of the other columns. If there are strong correlations among the explanatory variables, there will usually be approximate linear relations among the columns. The result is that the standard errors for the estimated regression coefficients may be drastically increased and thereby may even cause estimates of true coefficients with small magnitudes to have the wrong sign.

To avoid these problems, the common features of large bodies of aerometric data have been examined using alternative multivariate methods. Two such methods are regression on principal components, which eliminates problems with collinearity in explanatory variables, and empirical orthogonal function analysis (principal-component analysis) in which intersite covariations between end-product variables are examined. These methods also offer an efficient means of screening large amounts of data for internal consistency in the relationships between the explanatory variables.

Regression on Principal Components

Principal-component analysis (Mardia et al. 1979) applies an orthogonal transformation to the explanatory variables, which results in a new set of uncorrelated variables that are linear combinations of the original variables. The transformation matrix is the matrix of eigenvectors of either the correlation or the covariance matrix of the explanatory variables. The first principal component is the linear combination of variables that explains or accounts for the maximum variability in the original variables. The second principal component is that linear combination, uncorrelated with the first principal component, that explains the maximum amount of variability not already accounted for by the first component. The third, fourth, and other principal components are defined similarly. There are as many

principal components as there are linearly independent explanatory variables; however, the first few principal components usually account for most of the variability in the explanatory variables. The new variables or principal components have been interpreted in terms of physically significant factors such as chemical or transport processes (e.g., Henry and Hidy 1979).

When the principal components are found using the correlation matrix of the explanatory variables rather than the covariance matrix of the explanatory variables, the coefficients in the regression of end products on the new variables are "standard partial regression coefficients" (Snedecor and Cochran 1967). They provide a measure of the comparative "strength" of association between end-product variability and the new principal-component explanatory variables. The reason for this interpretation is that principal components are uncorrelated, so that a coefficient pertaining to one component is linearly unrelated to coefficients pertaining to the other components. In addition each component has unit variance so that the component is scale-free.

Considerable progress has been made in interpreting the temporal behavior of ambient sulfate by this type of approach. For example, Henry and Hidy (1979, 1982) analyzed large data sets from New York City, St. Louis, Los Angeles, and Salt Lake City using regression on principal components. In New York, St. Louis, and Los Angeles, the regression of sulfate on SO_2 at the same measurement location was not significant. Instead, the explanatory variables, ozone, temperature, and absolute humidity, accounted for the variability in sulfate data. In Salt Lake City, sulfate was found to be related to SO_2 concentrations and to conditions promoting atmospheric mixing. Evidently, variations in ambient SO_2 levels in themselves have only a weak influence on variations in airborne sulfate concentrations in these cities, compared with the influences of variations in atmospheric processes or meteorological conditions.

Rural data from the northeastern United States have also been examined using these techniques (Henry et al. 1980, Mueller and Hidy 1983). Regression on principal components of rural data indicated very complicated relationships between aerometric variables. However, significant evidence for a direct relationship between sulfate levels and ambient SO_2 concentrations of 3-hour and 24-hour averaged SURE data emerged from the analysis. With regression on principal components, a linear

proportionality between ambient SO_2 and ambient sulfate variability at the same site can be derived objectively. If regional SO_2 concentrations in air are proportional to SO_2 emissions and if dry deposition is proportional to ambient sulfur oxide concentrations at ground level, then one can infer that reduction in SO_2 emissions should logically result in reductions in dry deposition, even with the dominance of meteorological factors.

Empirical Orthogonal-Function Analysis

The influence of the spatial distributions of SO_2 emissions on ambient sulfate distributions also can be obtained using empirical orthogonal-function analysis to analyze sulfate measurements at a number of stations over a period of time (e.g., Henry et al., 1980, Mueller and Hidy 1983, Peterson 1970). This technique derives eigenvectors from the estimated covariances of sulfate concentrations between pairs of stations. Use of covariances rather than correlations gives greater weight to sites with larger sulfate variability. Spatial patterns are then derived for each empirical orthogonal function (or eigenvector) separately by plotting the value of the eigenvector for each station, interpolating the values onto a grid, and then drawing contours from this grid.

Examples of the first two empirical orthogonal functions derived from the SURE data for July 1978 are shown in Figure 4.12. The patterns shown are contours of constant values of the empirical orthogonal functions, where stations with the same sign covary together (when one is high, the other is high) and stations with opposite signs vary in opposition. The shading indicates areas of high SO_2 emission density.

The empirical orthogonal function that accounts for the largest amount of sulfate variability in the data, identified as the "first," is shown in Figure 4.12(a). The function accounting for the second largest amount of the variability, identified as the "second" function, accounts for the next largest fraction of sulfate variability [Figure 4.12(b)]. For the July 1978 data, the first two empirical orthogonal functions account for 74 percent of the deviation of the sulfate concentration from its mean value.

The spatial patterns of August 1977 were similar to the July 1978 patterns shown in Figure 4.12; however, the patterns obtained from the SURE data differ from season

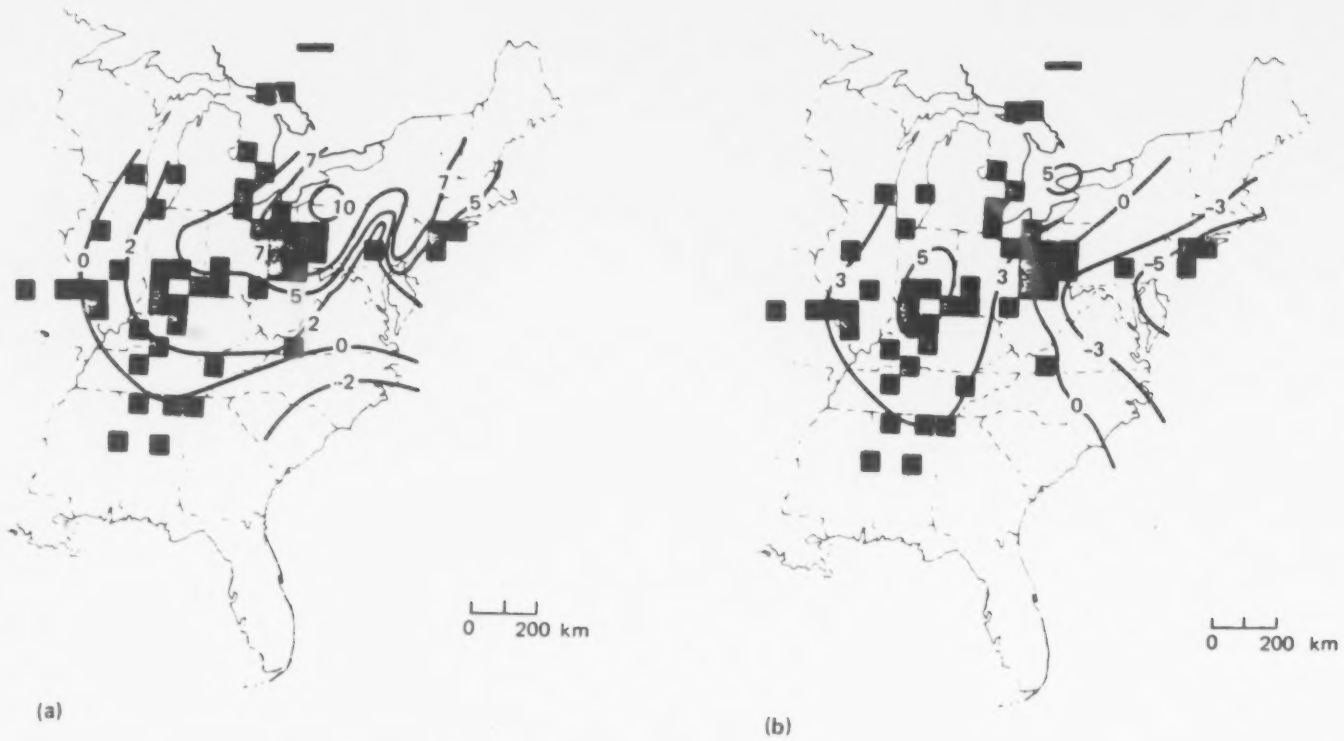


FIGURE 4.12 Empirical orthogonal functions (EOF) in the SURE data for July 1978. Shown are the lines of constant values of the EOFs in micrograms of sulfate per cubic meter. Shaded areas denote SO_2 emissions greater than 500 Mt/day. Of the total sulfate variance between locations where measurements were taken, the first EOF explains 54 percent (a), while the second EOF explains 20 percent (b). SOURCE: Henry et al. (1980).

to season (e.g., summer to fall or winter). In all of these cases, the patterns can be rationalized to be consistent with the dominant weather observed during the periods examined. In summer, for example, the pattern of the first empirical orthogonal function was identified with large-scale meteorological conditions leading to persistent mass transport of air around zones of high barometric pressure northeastward across the Ohio River Valley toward New England. These conditions were identified as being most likely for long-range pollution transport (Mueller and Hidy 1983). The second important empirical orthogonal function in summer was identified with large-scale air stagnation under conditions of poor ventilation on the west side of summer anticyclones (e.g., Vukovich et al. 1977). (It should be noted that there were 10 days with conditions of air stagnation during July 1978, whereas the 40-year average for July is 2 days.)

The association between SO_2 emission distributions and the important empirical orthogonal functions found in the SURE data is circumstantial evidence that changes in the source patterns should induce changes in regionwide sulfate variability over the eastern United States. However, empirical orthogonal-function analysis and other statistical techniques have not progressed to the stage at which they can be used to predict consequences of selective changes in emissions.

Elemental-Tracer Analysis

Sources often emit particulate matter that may serve as chemical or elemental tracers of the emissions. Sampling and analysis for the tracer substances at receptor locations can provide evidence of the influence of various types of sources on those locations. Because monitoring stations are in the receiving field, these statistical approaches to analyzing empirical data are often called receptor models.

The technique, developed originally for studies of urban air quality, has recently been proposed for application to long-range transport of acidic substances. The method relies on the fact that particles associated with different sources have different elemental compositions (Gordon 1980, National Research Council 1980). For example, sodium in particles collected in aerosol monitors is usually associated with

a marine source, such as salt from windblown sea spray. Other elements in particles that are strong indications of particular sources are calcium (limestone, hence construction and demolition activities), lead (motor vehicles), vanadium (fuel oil), and zinc (municipal refuse). Particles from coal combustion are more difficult to distinguish because their elemental composition is much like that of soil. However, coal combustion produces fine particles that are significantly enriched in selenium and arsenic with respect to soil, so these elements potentially can be used to distinguish the contributions of coal combustion from those of windblown dust. The tracer method has been tested and improved for application to urban air sheds in several U.S. cities, including Washington, D.C. (Kowalczyk et al. 1982), Los Angeles (Friedlander 1973, Gartrell and Friedlander 1975), Portland, Oregon (Core et al. 1981), and St. Louis (Alpert and Hopke 1981, Dzubay 1980). The method has also been applied on the scale of global circulation to determine the sources of pollution particles collected at sites remote from human activities, such as in the Arctic (Cunningham and Zoller 1981; Heidam 1981, 1982; Rahn 1981a,b).

It has been suggested (for example by K. Rahn, University of Rhode Island, in an unpublished paper, 1982) that the elemental composition of particulate matter in aerosols might also be indicative of the origins of the aerosols after regional-scale transport and might help to resolve the question of the relative contributions of distant and local sources to acid deposition in eastern North America. Among major stationary sources that contribute substantially to the total burden of emission of SO_2, those in the eastern United States are fueled by coal and oil, whereas those in the Midwest burn mainly coal. Thus an aerosol collected in the East that is enriched in vanadium would be presumed to be of relatively local origin, whereas an aerosol characteristic of coal combustion would be taken to be of midwestern origin.

As yet there has been no systematic evaluation of the method, nor has it been applied to a number of available sets of data. Care should therefore be exercised in interpreting the results of preliminary analyses using the method, which may be more difficult to apply on a regional scale than on either an urban or a global scale.

Urban air quality is usually determined by local emissions, so the elemental composition of particulate

matter collected at urban monitoring sites can be expected to reflect the types of sources in the urban air shed, with the possible exception of aerosol particles, such as sulfates, formed in the atmosphere from precursor gases and transported long distances. Polluted air reaching remote sites, by contrast, has usually undergone so much mixing that its elemental composition can be considered to be a composite representative of a large source area. When polluted air travels from the source region to a remote site mostly over the ocean, however, there are few additional sources contributing particles to the air mass during transit, and the method can give unambiguous results. At rural continental sites, the dominant local source of airborne particles is usually soil. Most other types of particles come from distant sources such as large power plants or major metropolitan areas.

There appear to be two critical problems in the application of elemental tracer analysis to regional transport that must be solved before the method can be useful. The first problem arises because of the potential that the elements of interest as tracers may not be transported at the same rates and over the same distances as sulfates and nitrates, which are of primary concern in acid deposition.

For example, manganese and vanadium have been proposed as characteristic tracers of emissions from midwestern and eastern source regions, respectively. Manganese, most of which arises from entrained soil, is typically found on relatively large particles that settle to the ground close to the source; only about one third of airborne manganese is found in fine particles that would be expected to undergo long-range transport. Elemental tracer analysis attempts to correct for the presence of crustal material. Because most of the crustal manganese occurs on large particles, the technique focuses on manganese in fine particles. The source of airborne fine-particulate manganese is uncertain; coal is deficient in manganese relative to soil, for example. Almost all of the vanadium in particulate matter in the East is associated with fine particles. Whether deposition processes for sulfates and nitrates are similar to those of manganese and vanadium is unknown. The elements used as tracers are normally emitted in the form of solid particles, whereas the precursor gases SO_2 and NO_x are transformed into particles by chemical processes after emission. To the extent that formation of sulfate and nitrate is dominated by in-cloud processes, it seems

likely that different processes will govern transport and deposition of the acids and the tracers. Coal combustion is predicted to be a major source of airborne selenium, appreciable fractions of which leave stacks in the vapor phase and condense on particles as the exhaust gases cool in the atmosphere. The behavior of selenium may therefore be more like that of sulfur in the atmosphere than is that of manganese.

The second, and perhaps more difficult, problem is that of differentiating contributions of midwestern sources from those of eastern cities over which an air mass originating in the Midwest may pass before it reaches sensitive receptor areas. While experience with the technique indicates that it is practical to differentiate the contributions to urban aerosols of types of sources in a local area, it may not be possible to assess the contributions to regional aerosol burdens of sources in different urban areas. It may indeed be possible to distinguish eastern air masses from midwestern air masses that are uncontaminated by emissions from eastern cities but not possible to identify a midwestern air mass that has passed over and received inputs from eastern sites. It seems likely that arrays of elements, rather than a single one, will have to be used as tracers of pollution of midwestern origin.

ANALYSIS OF HISTORICAL TRENDS

In this and the next section we analyze the relationships between emissions and wet deposition using data obtained from monitoring networks. The central question is whether the data can be used to judge whether deposition is linearly or nonlinearly related to emissions. (See Chapters 2 and 3 for discussions of the processes that might lead to a nonlinear relationship.) If the relationship is linear, a change in emissions would be reflected in a proportionate change in deposition; if it is nonlinear, the change in deposition, if any, would be disproportionate.

Data on precipitation for analysis are available from both Europe and North America. Considerably more data are available from Europe, where monitoring programs have been in existence for many years, than in North America, where systematic monitoring of precipitation chemistry is a comparatively recent development. In this section we use the body of historical data as direct evidence of the

relationships, and then, owing to the paucity of these data, we assess the indirect evidence for linearity or nonlinearity in North America in the next section. More precisely, we consider whether currently available observations are consistent with conditions that should theoretically prevail if the relationship between emissions of SO_2 and NO_x and wet deposition of sulfate and nitrate in North America were strongly nonlinear.

The possibility that changes in deposition rates are not linearly related to changes in source strength initially arose in the examination of historical trends in emissions and deposition in Europe. The body of European data provides the only direct evidence for this phenomenon. For example, from about 1960 to 1975, most of the areas in the European network for precipitation sampling experienced either constant or declining deposition of sulfate (Figure 4.13) despite significant increases in SO_2 emissions during that period. This apparent nonlinearity of response has been attributed both to chemical factors and to changing climatic conditions. The data analyzed by Granat (1978) were collected using the bulk sampling technique, by which collection containers continuously exposed to the atmosphere collect combined wet and dry deposition. The observed trends are made somewhat uncertain by changes in analytical techniques, sampling techniques, and analytical laboratories throughout the sampling period. Furthermore, bulk deposition samples are subject to contamination from windblown dust, leaves, and insects, for example, and consequently are considered to be of poorer quality than current methods, which collect only wet deposition.

Extensive direct evidence, comparable with that for Europe (Kallend et al. 1983), that can demonstrate a disproportionate relationship between emissions and deposition does not exist in North America.

Efforts to reconstruct historical trends from the sporadic and disparate data that have been collected in the United States since the late 1950s (Likens and Butler 1981) are beset by large uncertainties (Hansen and Hidy 1982, Stensland and Semonin 1982). The monitoring of precipitation chemistry at the Hubbard Brook Experimental Forest appears to provide the longest continuous record of deposition data at a receptor site in the northeastern United States. These data, as in the European network, were obtained from samples of bulk deposition. The Hubbard Brook samples were collected on a weekly basis,

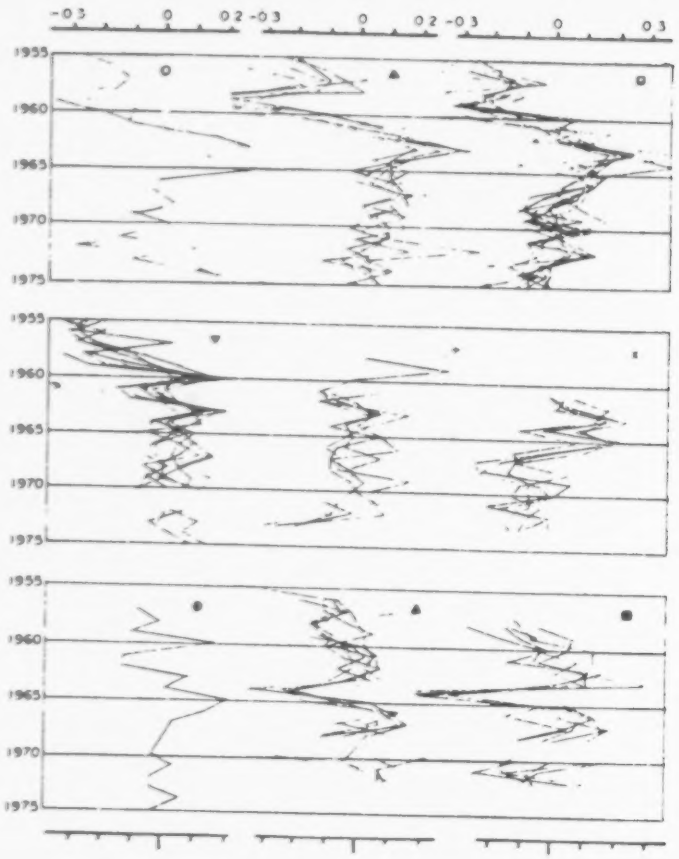


FIGURE 4.13 Long-term variation in deposition of excess sulfate for groups of stations in Europe. Symbols identify stations in each group. Scale is logarithmic. SOURCE: Granat (1978).

usually with three samples collected simultaneously. We regard the Hubbard Brook bulk-deposition data as reasonably reliable because samples were collected frequently and the collection of several samples at one site permits the detection and elimination of contaminated data.

The Hubbard Brook data reveal several trends (Likens et al. 1980), which are borne out at least qualitatively in bulk deposition monitoring with relatively unreliable quality control from nine stations in the New York State area (Miles and Yost 1982, Peters et al. 1982): (1) There has been a decrease in sulfate concentration since 1964 but an increase in nitrate concentration over the same time (Figure 4.14 and Table 4.4). (2) The annual pH of precipitation showed no long-term significant change from 1964 to 1977, though several short-term changes did occur. (3) A linear regression equation of data points from 1964 to 1977 indicates no statistically significant trends in H^+ deposition from 1964 to 1977. (4) Recent changes in H^+ deposition correspond more with changes in nitrate deposition than with sulfate deposition, even though sulfuric acid is the dominant acid at Hubbard Brook. The contribution of NO_3^- to total acidity has been increasing, whereas that of $SO_4^=$ has been decreasing (Galloway and Likens 1981). Year-to-year changes superimposed on the long-term trend may be related to climatological influences.

A linear-regression analysis of the Hubbard Brook data against time indicates a decline in sulfate concentrations between 1965-1966 and 1979-1980 of about 33 ± 18 percent (95 percent confidence limits). The regression of sulfate concentration against time gives a slope of -0.074 (standard error 0.016) and an intercept of 3.186 (standard error 0.138). Using the t test, the slope is significantly different from zero at the 0.0003 level. The assumptions of the regression--that errors on the fit are independent, have zero mean and a constant variance, and follow a normal distribution--were tested and do not appear to have been violated.

The Hubbard Brook record, unlike the European data, appears in toto to demonstrate a reduction in sulfate concentration similar to the general reduction in SO_2 emissions (Figure 4.15 and Table 4.5). Between 1965 and 1978 SO_2 emissions in EPA Region I (comprising Connecticut, Maine, Massachusetts, New Hampshire, Rhode Island, and Vermont) declined by about 38 percent (excluding data for Vermont, which are not reported in

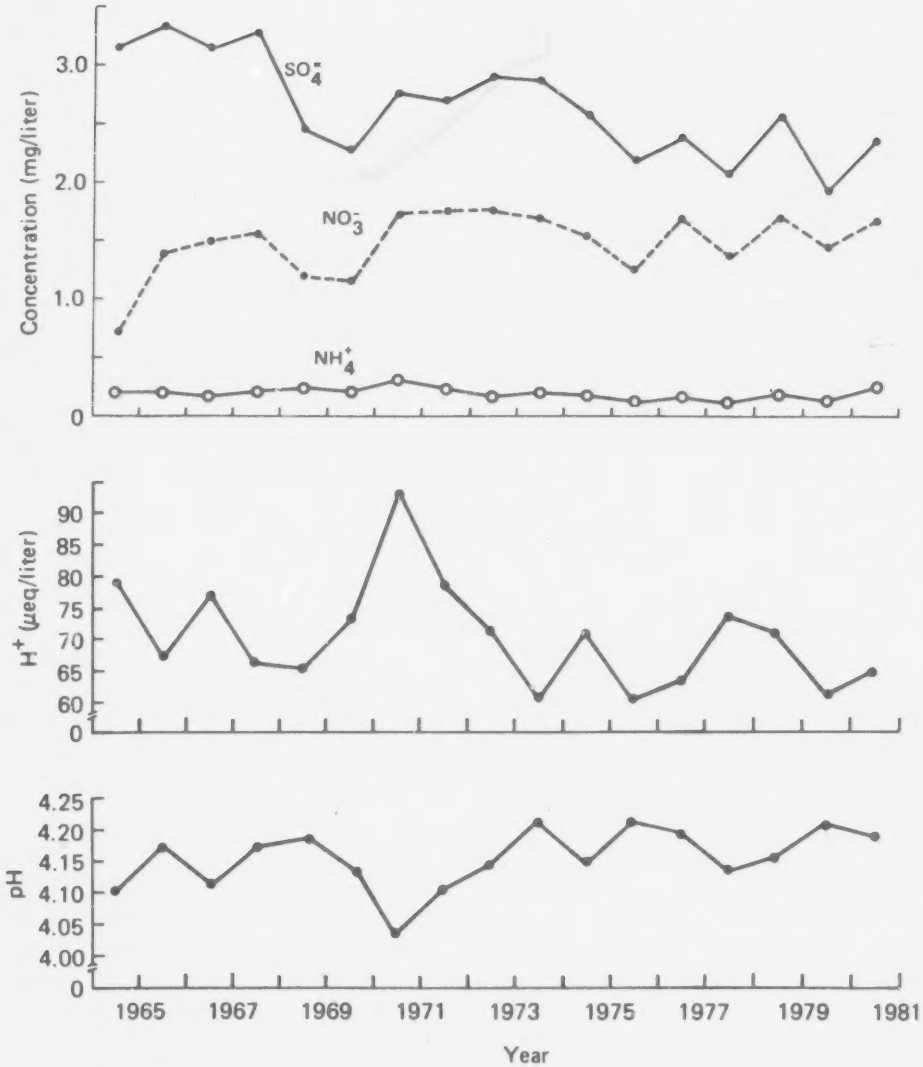


FIGURE 4.14 Annual weighted concentrations of sulfate, nitrate, ammonium, and hydrogen ions and weighted pH of precipitation at Hubbard Brook Experimental Forest from 1964 to 1977. SOURCE: Likens et al. (1980) and G.E. Likens, Cornell University, personal communication (1983).

the reference cited for Table 4.5). In Region II (New Jersey and New York) the reduction was about 40 percent, while that in Region III (Delaware, the District of Columbia, Maryland, Pennsylvania, Virginia, and West Virginia) was about 11 percent during the same period. Aggregate sulfur emissions in Region IV (Alabama,

TABLE 4.4 Annual Average Concentrations of Sulfate and Nitrate from Weekly Bulk Samples at Hubbard Brook Weighted by the Annual Amount of Precipitation (mg/liter)[a]

Year	$SO_4^=$	NO_3^-
1964/65	3.16	0.70
1965/66	3.33	1.39
1966/67	3.13	1.49
1967/68	3.27	1.56
1968/69	2.42	1.18
1969/70	2.24	1.14
1970/71	2.75	1.71
1971/72	2.67	1.74
1972/73	2.87	1.74
1973/74	2.84	1.67
1974/75	2.54	1.52
1975/76	2.14	1.22
1976/77	2.20	1.66
1977/78	2.04	1.32
1978/79	2.55	1.69
1979/80	1.91	1.44
1980/81	2.36	1.66

SOURCE: G.E. Likens, Cornell University, personal communication, January 9, 1983.

[a]Data are recorded in the water year, from June 1 through May 31.

Florida, Georgia, Mississippi, Kentucky, North Carolina, South Carolina, and Tennessee) increased by about 33 percent between 1965 and 1978. Region V emissions (from Illinois, Indiana, Michigan, Minnesota, Ohio, and Wisconsin) decreased by approximately 18 percent over the period. Total SO_2 emissions from EPA Regions I through V, which comprise all states east of the Mississippi River plus Minnesota, decreased by about 8 percent between 1965 and 1978.

The nitrate data at Hubbard Brook suggest an erratic trend toward a maximum around 1970, followed by a leveling off or a slight decrease. The emissions of NO_x in the Northeast increased 26 percent between 1960 and 1970, and then decreased 4 percent by 1978. The nitrate deposition data also appear to reflect emission trends in the Northeast.

The data of Peters et al. (1982) were obtained from a network operated by the U.S. Geological Survey (USGS). The USGS network employed bulk samplers; single samples were collected on a monthly basis.

The scatter in the USGS data is quite large. According to Peters et al. (1982), the data do not suggest statis-

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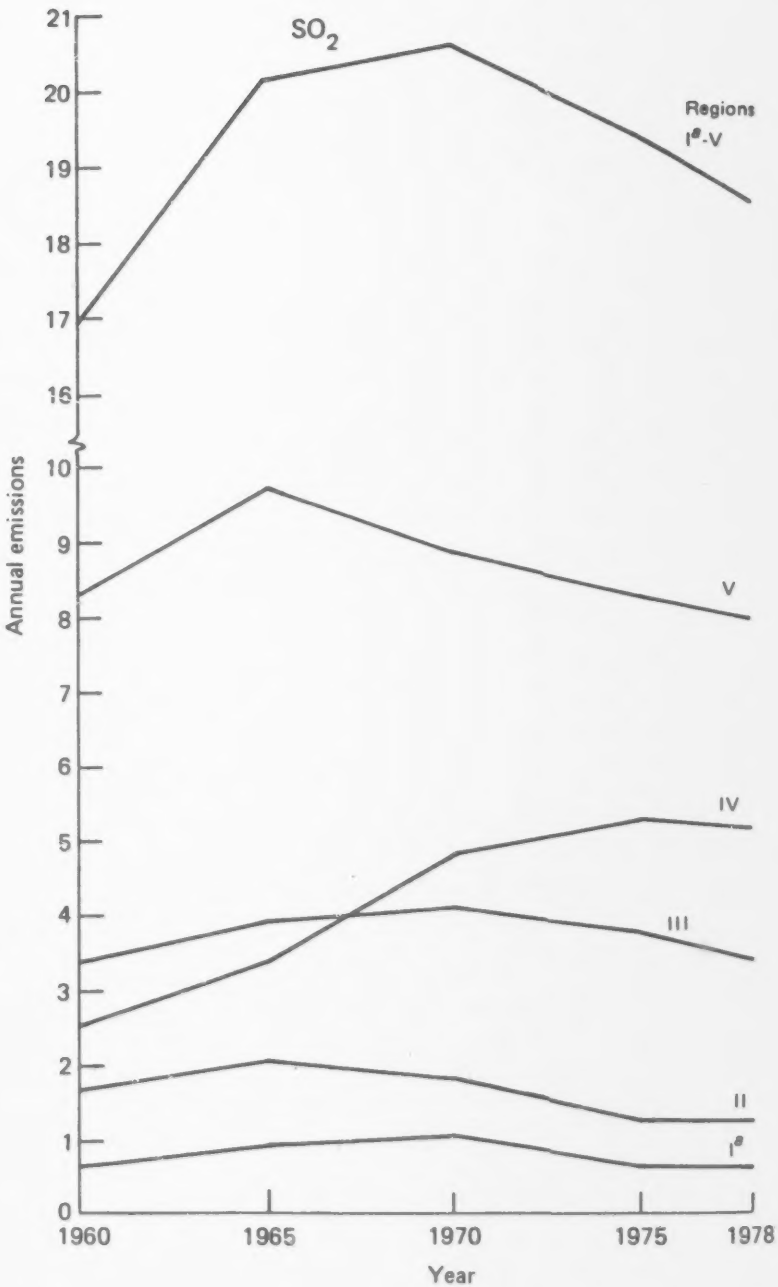

[a]Excludes emissions from Vermont, which in 1980 amounted to about 7,500 tonnes of SO_2 and 12,000 tonnes of NO_x.

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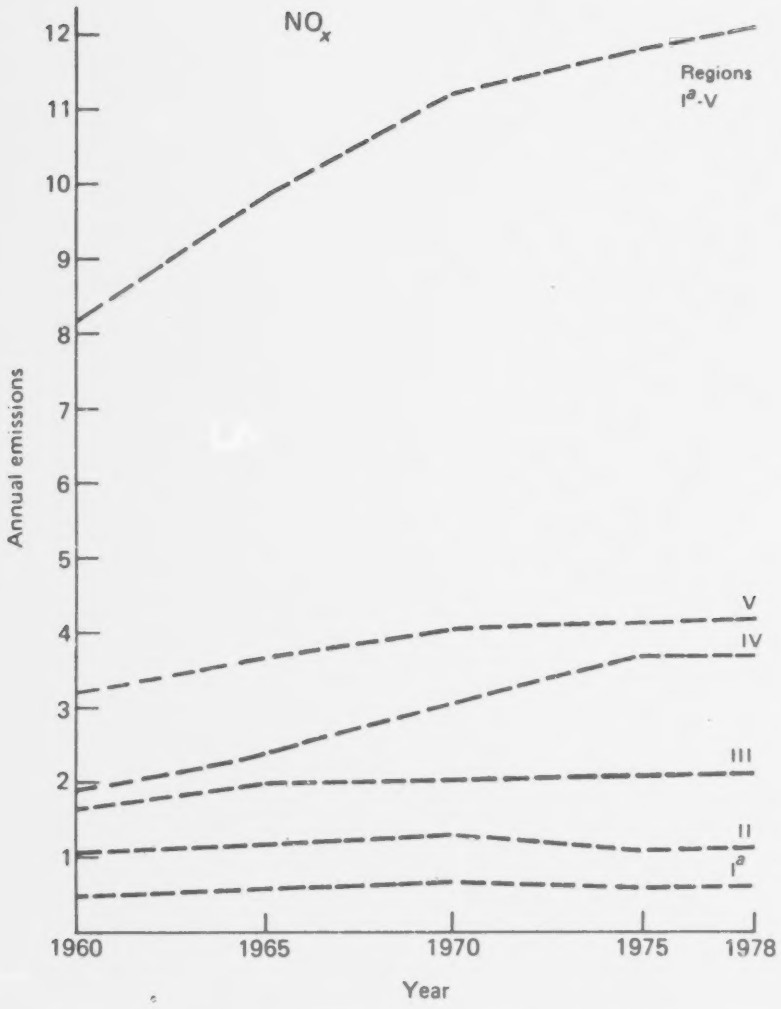

FIGURE 4.15 Annual emission of SO_2 and NO_X in the eastern United States by EPA Region, 1960-1978 (million metric tonnes). SOURCE: U.S./Canada Work Group #3B (1982).

tically significant long-term trends except for sulfate at one site (Hinckley). Although the direction of change is consistent, these results suggest weaker trends since the mid-1960s than the Hubbard Brook results. Uncertain quality control, especially during the early years of operation obviates the usefulness of these data however (Miles and Yost 1982).

Another source of data is the New York State Department of Environmental Conservation (1976, 1978,

TABLE 4.5 Annual Emissions of SO_2 and NO_x for Selected States Adjacent to or East of the Mississippi River, 1960-1978 (thousands of metric tonnes)[a]

State	SO_2					NO_x				
	1960	1965	1970	1975	1978	1960	1965	1970	1975	1978
Alabama	552	803	881	888	686	278	403	374	523	426
Connecticut	217	412	286	172	101	137	152	182	164	165
Delaware	177	196	201	174	169	46	55	65	59	64
District of Columbia	35	43	70	24	16	32	34	52[b]	33	30
Florida	307	451	776	745	617	289	379	497	660	700
Georgia	178	273	369	514	636	204	267	358	469	494
Illinois	2,208	2,512	2,256	1,756	1,573	806	957	1,008	1,016	1,017
Indiana	1,657	1,962	1,747	1,782	1,663	526	500	519	569	541
Iowa	328	397	333	283	347	195	223	279	278	289
Kentucky	568	823	1,153	1,320	1,197	251	340	447	511	507
Maine	63	87	74	61	59	44	54	68	65	69
Maryland	466	529	421	290	322	201	263	269	265	283
Massachusetts	337	399	526	326	362	229	273	324	306	328
Michigan	977	1,370	1,369	1,306	1,006	529	672	762	757	757

Minnesota	353	378	406	344	341	216	248	298	333	360
Mississippi	37	40	72	174	238	136	177	274	219	246
Missouri	524	607	997	1,057	1,177	265	305	382	534	507
New Hampshire	26	37	86	68	61	28	36	57	61	60
New Jersey	434	561	531	307	291	326	395	484	416	445
New York	1,285	1,481	1,310	971	937	690	827	900	782	818
North Carolina	209	265	480	451	506	261	339	492	511	532
Ohio	2,640	2,863	2,813	2,944	2,804	864	979	1,049	1,099	1,149
Pennsylvania	2,126	2,292	2,021	1,918	1,710	918	1,029	980	984	1,009
Rhode Island	79	37	55	22	18	41	33	50	40	38
South Carolina	104	110	167	182	260	135	160	214	228	270
Tennessee	658	694	889	1,028	1,047	302	342	420	554	534
Virginia	154	169	428	343	324	234	326	390	379	392
West Virginia	477	699	882	1,098	945	203	290	312	424	416
Wisconsin	544	633	290	150[b]	597	267	331	410	410	426

SOURCE: U.S./Canada Work Group #3B (1982).

[a]Data on emissions in Vermont for this period are not contained in the referenced source and are believed to be small.

[b]Questionable datum.

TABLE 4.6 Three-Year Running Average Values for Sulfate Concentrations in New York ($\mu g/m^3$)

Year	New York Statewide Sulfate Levels	Rochester Station 2701-01
1964	10.78	
1965	11.01	
1966	10.68	9.6
1967	9.88	9.37
1968	9.46	9.40
1969	9.04	8.83
1970	8.89	8.23
1971	8.84	8.13
1972	9.01	8.70
1973	9.00	8.77
1974	9.15	8.57
1975	9.32	8.53
1976	9.45	9.40
1977	9.02	9.8
1978	8.72	10.0
1979	8.3	9.53
1980	8.42	9.5
1981	8.42	9.65

SOURCE: Department of Environmental Conservation (1976, 1978, 1981).

1981). According to these data, statewide concentrations of sulfate particles in New York have also decreased about 22 percent between 1964 and 1978 (Table 4.6). This decrease corresponds fairly closely to the decrease in Northeast SO_2 emissions (Figure 4.15). However, several stations, e.g., at Rochester, reveal more complex trends throughout the period (Table 4.6). These data are difficult to interpret. Many of the stations are located in urban areas so that both the statewide average and values from specific stations should be strongly influenced by local urban sources. In addition, controls for both SO_2 and primary sulfate emissions were implemented during this period.

Both the USGS and New York State data are therefore of limited usefulness for assessing relationships between emissions and deposition influenced by long-range transport to rural areas. The most reliable data available in a continuous record are those from Hubbard Brook. These data comprise the direct historical evidence for an emissions-deposition relationship and show no indication of a significant nonlinearity.

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ANALYSIS OF RELATIVE BEHAVIOR OF SULFUR AND NITROGEN EMISSIONS

Because of the small amount of direct evidence available, we resort to examining less direct empirical evidence that may reveal for eastern North America a nonlinear relationship between emissions and deposition.

An explanation of the inconsistent emission and precipitation trends observed in Europe comes from work of Rodhe et al. (1981), who hypothesized that (1) gas-phase photochemistry is significantly responsible for producing the acid incorporated in precipitation and (2) conversion of SO_2 to H_2SO_4 is indirectly influenced by emissions of NO_x in the atmosphere (see the detailed discussion in Chapter 3 on the Rodhe et al. model). In the Rodhe hypothesis, HNO_3 production is thought to be favored near the source region, while H_2SO_4 is favored in more distant regions. Nitric acid vapor formed by the reaction of NO_x and the OH radical can be rapidly removed from boundary-layer air by either dry deposition (B.B. Hicks, National Oceanic and Atmospheric Administration, private communication, 1982; Huebert 1982) or by wet deposition (Levine and Schwartz 1982). If the mechanisms suggested by Rodhe et al. significantly influence the concentrations of $SO_4^=$ and NO_3^- in precipitation, the ratio of dissolved sulfate to nitrate is expected to increase as the SO_2 oxidation becomes more efficient with increasing distance from the source region.

The occurrence of the gradient of ratios does not, however, unambiguously demonstrate the importance of this mechanism, since other factors (such as very rapid dry deposition of nitric acid vapor in relation to that of $SO_4^=$ aerosol) might yield a similar effect if gas-phase chemistry plays a dominant role. However, the absence of the gradient of ratios would be difficult to reconcile with the proposed model. Molar ratios of sulfate to nitrate in precipitation in Europe calculated from the data of Rodhe et al. (1981) do indeed increase with increasing distance from the source region in summer (Figure 4.16 and Table 4.7), when the influence of gas-phase chemistry is expected to be greatest.

The annual average of the molar ratio of sulfate to nitrate in precipitation is relatively constant throughout the area of the northeastern United States and eastern Canada in regions in which the pH precipitation is low but increases in extreme northern Canada (Figure 4.17). High values of the ratios in northeastern Canada

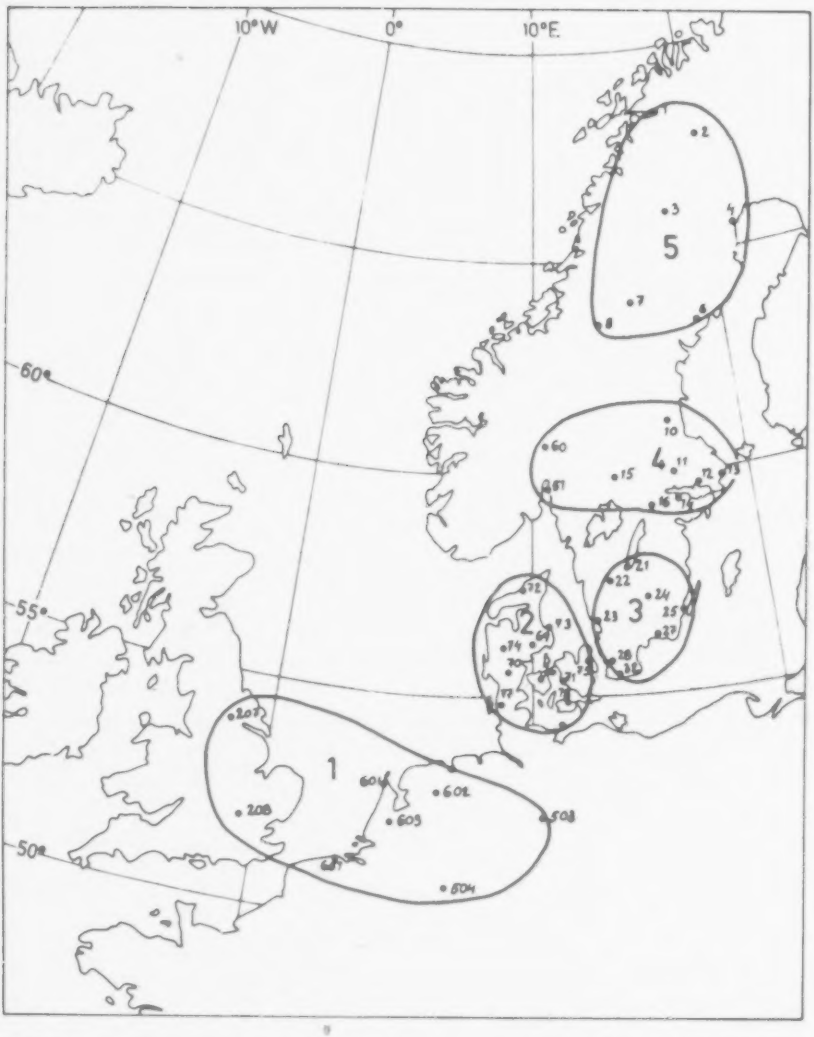

FIGURE 4.16 Locations of monitoring stations used in the data analysis of Table 4.7. SOURCE: Rodhe et al. (1981).

and far north Europe (region 5 in Figure 4.16) may be affected by local smelting facilities. Enhancements of the ratio occur in areas of concentrated point-source emissions, e.g., the Ohio and Tennessee River Valleys (Figure 4.18) and along coastal areas, possibly because of sea-salt sulfate. A lower ratio occurs near the Great Lakes, a region with a relatively low ratio of SO_2 to NO_x emissions. That the ratio is relatively constant over a broad region appears inconsistent with the exis-

TABLE 4.7 Sulfate, Nitrate, and the Molar Ratio of Sulfate to Nitrate in Precipitation in Europe

	Group of Stations[a]				
	1	2	3	4	5
Sulfate (μmoles/liter)					
1955–1959	62.5	50.0	28.1	25.0	15.6
1970–1974	90.6	56.3	50.0	40.6	40.6
Increase (%)	45	13	78	63	160
Nitrate (μmoles/liter)					
1955–1959	35.0	22.9	13.6	7.1	4.3
1970–1974	60.7	51.4	25.0	21.4	7.1
Increase (%)	73	125	84	200	67
Molar ratio					
1955–1959	1.8	2.2	2.1	3.5	3.6
1970–1974	1.5	1.1	2.0	1.9	5.7

SOURCE: Rodhe et al. (1981).

[a]Geographic locations of the groups of stations are indicated on Figure 4.16.

tence of a strong nonlinearity due to photochemistry and demonstrates the apparent effectiveness of atmospheric processes to lead to thorough mixing of pollutants over a large spatial scale (up to 1,000 km in linear dimension).

When the ratio of sulfate to nitrate in precipitation is considered on a monthly basis, the pattern becomes more complex. Average ratios have been calculated for the MAP3S stations (MAP3S/RAINE Research Community 1982) for two seasonal periods, April to September and October to March, for the period 1977 to 1981. The seasonal and annual ratios are given in Table 4.8. The ratio tends to be larger in summer than in winter, with the seasonal difference diminishing at lower latitudes (possibly because the intensity of sunlight varies less with season) or in maritime environments (where sea-salt sulfate may contribute to the ratio). The seasonal change may reflect a less efficient conversion of SO_2 than of NO_x during the cooler months than during the warmer months, when photochemistry is more active and transport is more vigorous. Incorporation of nitrate into precipitation is much less sensitive to seasonal change. Emissions of SO_2 are relatively constant throughout the year (16 percent increase in winter) (Electric Power Research Institute 1981). Seasonal variations in emissions, therefore, cannot account for the seasonal change in deposition ratios. The deposition ratios, however, are relatively invariable with respect to distance from major source regions, particularly during the warm season (1.4

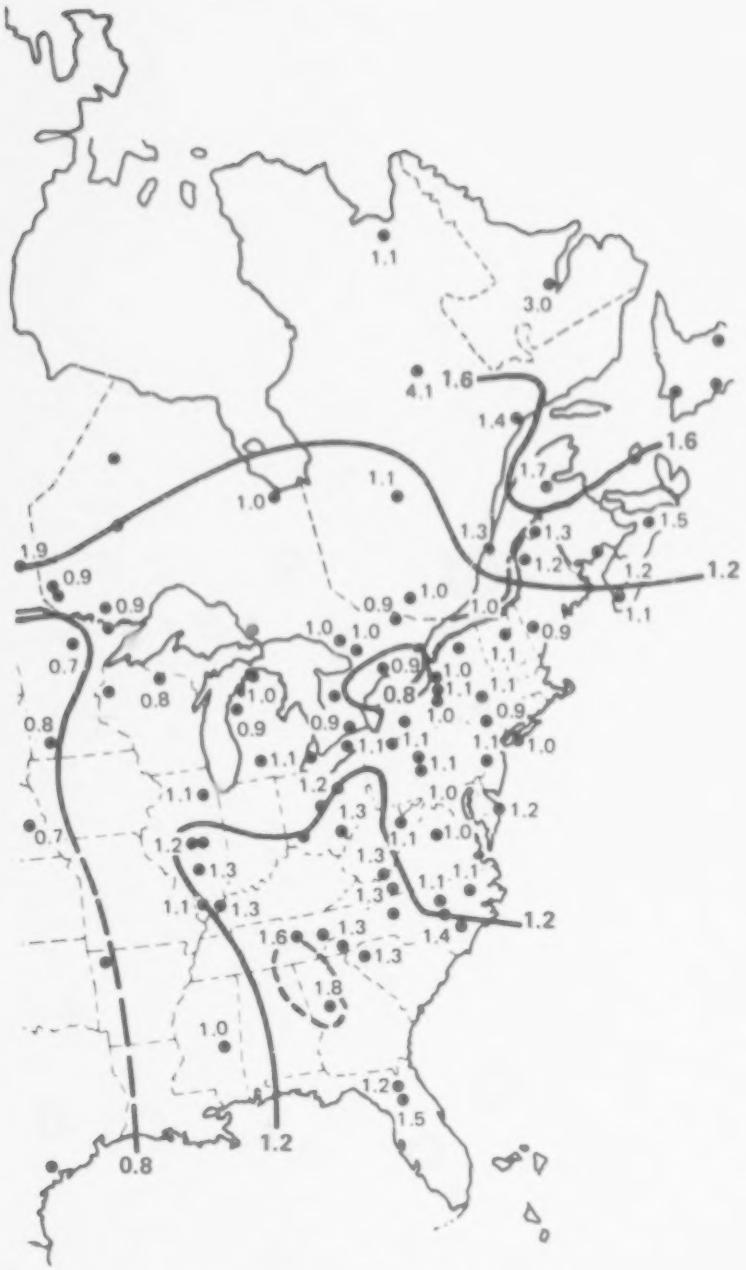

FIGURE 4.17 Average molar ratio of sulfate to nitrate in precipitation in eastern North America in 1980. SOURCE: B. Heikes, National Center for Atmospheric Research, personal communication (1983). Based on data from U.S./Canada Work Group #2 (1982).

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FIGURE 4.18 Average molar ratio of sulfur to nitrogen oxides in emissions in eastern North America in 1980. SOURCE: B. Heikes, National Center for Atmospheric Research, personal communication (1983). Based on data from U.S./Canada Work Group #3B (1982).

TABLE 4.8 Molar Ratios of Sulfate to Nitrate in Precipitation in the United States, 1977–1981[a]

Station	April–September		October–March		Annual Average
Whiteface Mt., N.Y.	1.4	(29)	0.65	(26)	1.08
Ithaca, N.Y.	1.4	(25)	0.62	(21)	1.07
University Park, Pa.	1.3	(27)	0.74	(29)	1.04
Charlottesville, Va.	1.3	(27)	0.81	(21)	1.16
Urbana, Ill.	1.4	(15)	0.94	(13)	1.25
Brookhaven, N.Y.	1.12	(17)	1.0	(17)	1.10
Lewes, Del.	1.4	(18)	0.93	(18)	1.16
Oxford, Ohio	1.5	(16)	1.2	(19)	1.37
Average	1.4 ± 0.1		0.86 ± 0.19		

SOURCE: Adapted from MAP3S/RAINE Research Community (1982).
[a]Numbers in parentheses are the number of months of data in each sample.

± 0.1) when gas-phase photochemistry would be expected to exert its maximum impact on rain composition in accordance with the model of Rodhe et al. (1981). In neither season do the ratios significantly increase with distance from the most highly industrialized areas, suggesting that retardation of sulfate formation by NO_x may be a minor factor at any time of the year in eastern North America.

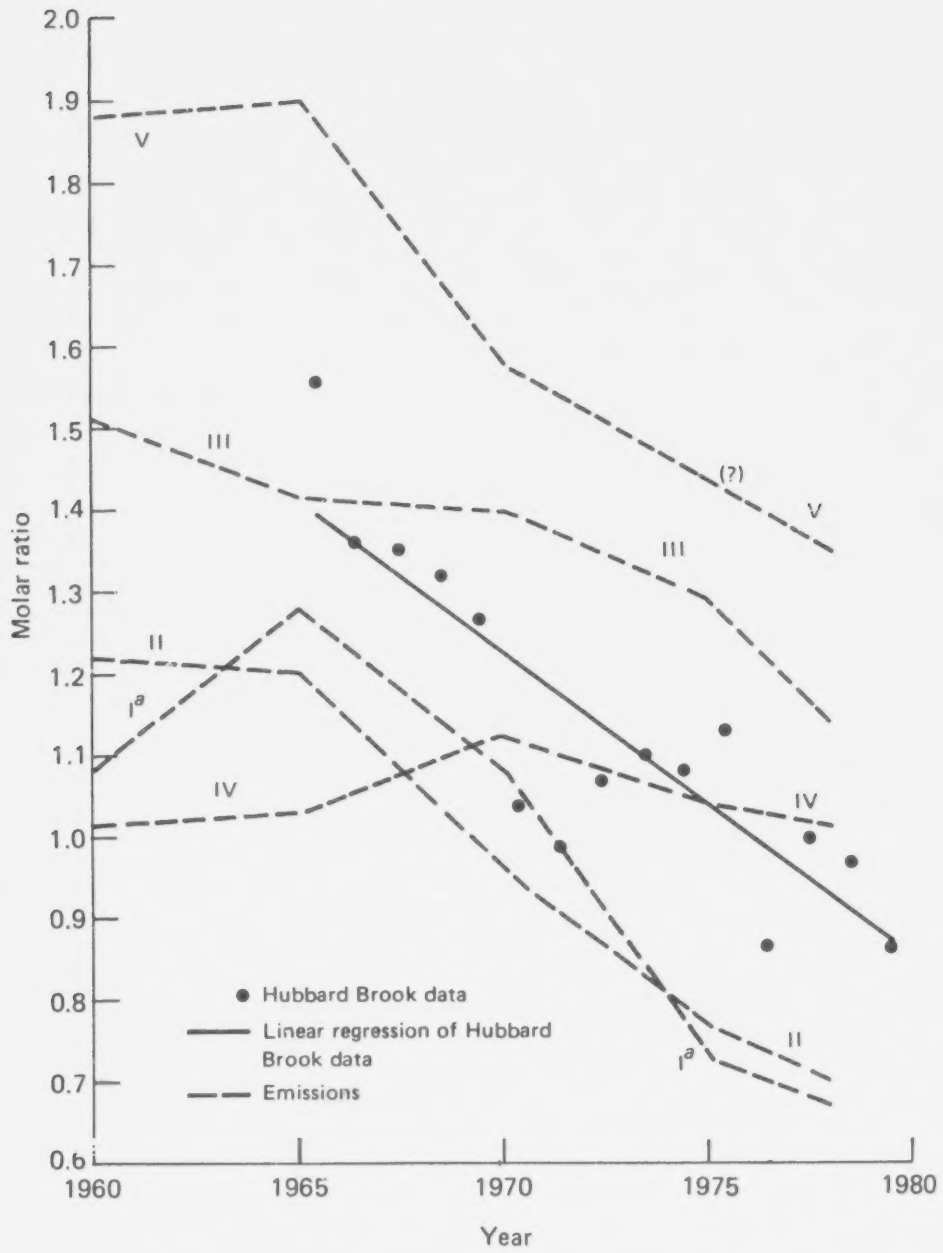
Accounting for the surprising relative invariability of the ratio despite the geographic variability of conditions requires considerable speculation. Perhaps the factor most responsible for uniformity is atmospheric mixing. Much of the frontal and convective precipitation in the eastern United States is from clouds forming in moist southwesterly air. Air-parcel trajectories have shown that this air is frequently advected several hundreds of kilometers before precipitation occurs (Lazrus et al. 1982, Raynor and Hayes 1982b). During this transit, the air incorporates and integrates the emissions from all the sources in its path. Another possibility is an unknown coupling of the NO_x and SO_2 chemical reaction pathways, which could tend to maintain a relatively unchanging deposition ratio of sulfate to nitrate, although there is no theoretical or experimental evidence of such a coupling.

It is surprising to note in addition that the annually averaged molar ratios in precipitation at the MAP3S, NADP, and CANSAP sites in eastern North America resemble the molar ratios of SO_2 to NO_x emissions averaged on an annual basis by state or province (Figure 4.18). The emission ratios are somewhat higher than the deposition

ratios in the Ohio and Tennessee River Valleys. Similarly, the emission ratios are somewhat lower than the low deposition ratios immediately north of Lake Ontario. The smaller spatial variability of the deposition ratios compared with the emission ratios near large point sources is probably related to a rapid rate of atmospheric mixing relative to the rates of wet-deposition processes. Alternatively, a preferential dry deposition of SO_2 relative to that of NO_2 may occur in the near-source region, or the differences may indeed represent a suppression of SO_2 oxidation within a short distance from the emission sources. However, the general similarity of wet-deposition and emission ratios on an annual average basis suggests that the incorporation of sulfuric acid in rain is not retarded with respect to the conversion and incorporation of nitric acid in rain.

A small body of data suggests that the historical trends in the molar deposition ratios of sulfate to nitrate correspond to the trend in molar SO_2/NO_x emission ratios. The historical trends of the molar deposition ratio $SO_4^=/NO_3^-$ at Hubbard Brook and the molar ratios of SO_2 to NO_x in emissions in the eastern United States by EPA Region are plotted in Figure 4.19. Also shown in the figure is the result of a linear-regression analysis of the Hubbard Brook deposition ratio against time. Between 1965-1966 and 1979-1980, the linear regression gives a slope of -0.039 (standard error 0.006) and an intercept of 1.444 (standard error 0.054). The significance level of the slope is 0.0001.

Another perspective is provided by comparing the annually averaged regional wet-deposition rates with regional emissions. Integrating wet deposition over the northeast quadrant of the United States [using the isopleths of Pack (1980) based on MAP3S and SURE data], Golomb (1983) found sulfate deposition to be 4.3×10^{10} (±20 percent) moles/year and total nitrate deposition to be 3.9×10^{10} (±25 percent) moles/year. Golomb calculated regional emissions from U.S./Canada (1981) to be 2.2×10^{11} moles/year of SO_2 and 1.8×10^{11} moles/year of NO_x. The emissions were similarly distributed geographically. Golomb also indicates that 19 ± 6 percent of the sulfur and 20 ± 7 percent of the nitrogen emitted in this region are wet deposited there. It is interesting to note that Golomb found the molar ratio of sulfate to nitrate wet deposited annually in this region to be 1.1, compared with an annually averaged molar ratio of emissions for SO_2 and NO_x of



[a] Excludes Vermont.

FIGURE 4.19 Molar ratio of SO_2 to NO_x in emissions in the eastern United States by EPA Region and molar ratio of sulfate to nitrate in deposition at Hubbard Brook, New Hampshire, 1960-1978. SOURCE: Adapted from Tables 4.4 and 4.5.

1.2. The similarity of the ratios suggests that, whatever specific processes are taking place, there is a tendency on an annual basis for sulfate and nitrate to be deposited in the northeast quadrant of the United States with no significant loss of one component relative to the other.

Two inferences result from this observation. On an annual basis there is no indication that within the region the oxidation of SO_2 is retarded relative to the oxidation of NO_x. Laboratory evidence, field observations, and photochemical theory predict a relatively rapid gas-phase oxidation of NO_x to HNO_3, especially in summer (see Appendix A). If the rate of SO_2 oxidation (as a result of both gas-phase and aqueous reactions) is comparable with that of NO_x, then no significant retardation of sulfate formation is likely to occur within the eastern region of high acidity as a result of limited availability of gaseous or aqueous oxidants.

On the basis of Golomb's calculation and our own analysis, we conclude, therefore, that (1) on an annual average basis there is no evidence that the hypothesis of Rodhe et al. (1981) applies in the northeastern United States and further that (2) the net oxidation of SO_2 leading to sulfate in precipitation is as efficient as the net oxidation of NO_x leading to nitrate in precipitation in this region.

The first conclusion relates to a specific mechanism; the second to the core of the critical issue of nonlinearity. If SO_2 oxidation were limited by the availability of oxidant, it would be possible for SO_2 emissions to overwhelm, or saturate, the capacity of the atmosphere to oxidize all or most of the SO_2 within the region of eastern North America. Oxidation over the ocean and in remote land regions would presumably convert the remaining SO_2. Since a large fraction of emitted SO_2 could conceivably be in excess of the oxidant available in North America, a significant reduction in SO_2 might not yield a significant reduction in wet deposition of sulfate in North America, including, of course, the regions deemed sensitive to acid deposition. The second conclusion, however, suggests that the oxidative capacity is not saturated in the northeast. Furthermore, the similarity of deposition ratios in eastern Canada both to those in the eastern United States and to emission ratios upwind also tend to support this implication. The indirect evidence agrees with the small amount of reliable North American historical data, which

suggest that there is no evidence of a strong nonlinearity in the relationship between $SO_4^=$ deposition and SO_2 emissions.

The inference to be drawn from these data is that if the average ratio of SO_2 to NO_x in emissions in the region of high acidity covered by the data (consisting of the northeast quadrant of the United States, bounded by the southwest corner of Tennessee, the northwest corner of Illinois, New Hampshire, and the southeastern corner of North Carolina) were changed by changing SO_2 emissions while keeping NO_x emissions constant, that change should be reflected in a similar change in the ratio of sulfate to nitrate in wet deposition, all other emissions and conditions remaining unchanged. If dry deposition is linearly proportional to emissions, as suggested in the section on principal-component analysis, then the annual average ratio in bulk deposition in the area should also respond to changes in the emission ratio. The analysis is limited in its applicability to circumstances in which the spatial distribution of emissions is unchanged. Without greater confidence in the results of deterministic models, we cannot judge the consequences of emission reductions in a smaller region, such as the Midwest, for deposition in another region, such as the Adirondacks or southern Ontario (Chapter 3).

It is now necessary to consider the uncertainties inherent in the application of these observations to the problem of nonlinearity.

Our present inability to model the processes linking air composition with the composition of precipitation is revealed by a comparison of wet-deposition ratios with the molar ratios of SO_x to NO_x observed in air by the SURE network (Table 4.9). Precipitation scavenges airborne material both in and below clouds at some time after the injection of emissions into the atmosphere. Molar ratios in precipitation are expected therefore to reflect concentrations in ambient air. The ratios observed in ambient air at ground level in the SURE network, however, are not similar to those either in precipitation or in emissions. In ambient air at ground level, SO_x is depleted with respect to NO_x in the sum of gas and particle phases but is enriched in particles alone. The depletion of SO_x in ambient air relative to NO_x may be related to inherent nonlinearities in the processes involved, to differences in dry-deposition rates, to differences in the composition of air at ground and cloud levels, or to the comparison of ambient

TABLE 4.9 Molar Ratios of SO_x to NO_x in the Region of the SURE Experiment in 1977–1978[a]

Emissions	Ambient Air[b]		Precipitation
1.2 (all sources)	Gas and particles	0.66	0.73 to 1.3
2.2 (utilities)[c]	Gas only	0.42	
	Particles only	24.0	

SOURCE: Adapted from Mueller and Hidy (1983).

[a]Ratio taken as SO_2/NO_2.

[b]Sampled at ground level.

[c]Weighted heavily toward emissions injected at altitudes at or above 300 m.

concentration data that are averaged over all meteorological conditions with precipitation data that reflect only those conditions specifically favoring precipitation.

As indicated earlier, the ratio of sulfate to nitrate in precipitation varies regularly throughout the year, being high in summer and low in winter (see Table 4.8). It is unclear how the specific processes controlling these variations average out annually to be so uniform spatially and so similar to the emission ratios over much of the area when ambient atmospheric conditions apparently are so variable.

Conceivably the uniformity of the annual $SO_4^=/NO_3^-$ ratio in precipitation in the northeastern United States could reflect some as yet unidentified stoichiometric linkage between the atmospheric chemistry of SO_2 and NO_2. For example, the reaction of $HOSO_2$ radicals formed in reactions of HO with SO_2 could lead to $HOSO_2O_2$ and $HOSO_2O$ radicals, which could ultimately react preferentially with NO_2 to form a one-to-one, S to N compound such as $HOSO_2ONO_2$ (see Chapter 3 and Appendix A). This compound would in principle hydrolyze in cloud water to form equal amounts of H_2SO_4 and HNO_3 acids. However, this reaction mechanism would have to be the dominant source of $SO_4^=$ and NO_3^- to maintain the ratio of moles of $SO_4^=$ to moles of NO_3^- near unity, as observed in precipitation in the eastern United States. There is no laboratory or field evidence of the significant participation of nitryl sulfuric acid ($HOSO_2ONO_2$) or other similar compounds in acid development; in fact, the existing limited evidence appears to discount the possible importance of such a product as an intermediate in the HO-SO_2-NO_x reaction system (see Appendix A).

From theoretical considerations (e.g., Chapter 2 and Appendix A), we expect that ambient concentrations of oxidants (such as H_2O_2, O_3, and HO) will have

maximum values in summer and minimum values in winter. It is expected, therefore, that in winter in areas at higher latitudes, aqueous oxidation is slow because of low concentrations of oxidants in cloud water. Hence, if a limitation of oxidant were to lead to nonlinearity in the relationship between emissions and deposition, that effect would be most likely to be observed in winter. The evidence that the relationship between average annual emissions and deposition is not strongly nonlinear may result from the fact that most of the total annual deposition of acid occurs during the warm months.

Comparison of emission ratios and deposition ratios constitutes indirect evidence for the absence of a serious nonlinearity even though we lack an adequate understanding of the intermediate chemical and meteorological processes to predict, either qualitatively or quantitatively, the deposition ratios from the emission ratios. It is our opinion that the necessary data and theoretical understanding to model the effects of emissions from distant sources on the composition of precipitation reliably and accurately will not be available in the near future. The evidence based on the empirical observations appears incompatible with a seriously nonlinear system in terms of our current knowledge. Unquestionably, however, it would be preferable to reinforce such conclusions with a more complete understanding of atmospheric chemistry and meteorology under conditions prevalent in eastern North America.

Additional uncertainties are introduced by the inherent variabilities of natural processes (e.g., annual rates of precipitation), imprecision of measurements, and errors in determining emission rates. For example, the ratios of emissions and wet depositions in the Northeast are uncertain by a factor of about 30 percent. The concentrations of sulfate and nitrate in the MAP3S study are estimated to be uncertain by a factor of 15 to 20 percent.

A significant reduction in the uncertainties related to natural variability, measurement imprecision, or establishment of emission rates is unlikely in the near future.

FINDINGS AND CONCLUSIONS

Although data from which to assess the relationships between emissions and deposition are relatively sparse in

North America, analysis of the available empirical data provides insight into the nature of the atmospheric proceeses involved in acid deposition.

Nonlinearity

Data indicate that the variability in ambient concentrations of SO_2 vapor and $SO_4^=$ particles do not necessarily correlate with the variability in SO_2 emissions but are predominantly controlled by meteorological conditions. Variations in sulfate particle concentrations tend to correlate with variations in SO_2 vapor concentrations in rural areas of the Northeast. Regression on principal components and empirical orthogonal-function analysis suggest that if other factors were constant, ambient concentrations of SO_2 and $SO_4^=$ would be determined largely by patterns of SO_2 emissions.

Direct evidence of a strongly nonlinear relationship between wet deposition of sulfate and SO_2 emissions is limited to extensive historical data in Europe. The continuous historical record of reasonably reliable data in North America, at Hubbard Brook, indicates no evidence for a strongly nonlinear relationship between annual depositions and annual emissions in the Northeast. Indirect evidence based on patterns of the ratio of $SO_4^=$ to NO_3^- in annual deposition do not support the hypothesis of a strongly nonlinear relationship between SO_2 emissions and sulfate deposition in eastern North America. Differences in the relationship between emissions and deposition in Europe and eastern North America may be the consequence of differences in meteorology, latitude, or other factors, such as the spatial distribution of sources, between the two regions.

On the basis of currently available empirical data and within the limits of uncertainty associated with the data and with estimating emissions, we therefore conclude that there is no evidence for a strong nonlinearity in the relationships between long-term average emissions and depositions in eastern North America.

The conclusion that nonlinearity is probably not significant for annual average deposition in eastern North America is clouded by three types of uncertainties. First, direct evidence based on long-term time-series data is severely limited in North America to only ten stations, all of which have collected bulk deposition

data. The data from only one site, Hubbard Brook, are considered to be reasonably reliable. Therefore we have relied on the historical record at only one station combined with the indirect evidence provided by data on sulfate and nitrate deposition compared with SO_2 and NO_x emissions. Second, there remain uncertainties in our detailed understanding of the meteorological, physical, and chemical processes that relate emissions to deposition. Third, the unknown influences natural variability in the composition and occurrence of precipitation, imprecision in sampling and analysis, and uncertainties in estimation of emissions further limit confidence in this conclusion.

Influence of Local and Distant Sources

Both observational and theoretical evidence exists for the long-range transport of pollutants leading to acid deposition. It is apparent that any receptor site will be influenced to one degree or another by both local and distant sources. The issue of concern is the extent of this zone of influence for sensitive ecological areas, including the relative contributions to deposition of nearby and distant sources.

In the case of the Hubbard Brook data, the trends in concentrations of sulfate and nitrate appear to reflect general trends in emissions. Analyses of the trajectories of precipitating systems delivering acidic precipitation to Whiteface Mountain and Ithaca in New York and south central Ontario in Canada indicate that most of the acidity in precipitation at these sites--as well as most of the precipitation--is associated with air masses that have passed over source regions to the south and southwest.

The spatial distribution of the annual average molar ratios of pollutants in emissions and deposition suggest that atmospheric processes in eastern North America lead to a thorough mixing of pollutants over a wide geographic area, making it difficult to distinguish between the effects of distant and local sources.

On the basis of currently available empirical data, we cannot in general determine the relative importance for the net deposition of acids in specific locations of long-range transport from distant sources or more direct influences of local sources. We regard the problem of relating emissions from a given region to depositions in

a given receptor region to be of primary importance and recommend that high priority be given to research relevant to its solution.

The SURE data have indicated a relatively small zone of influence (of the order of 300 to 600 km) on ambient sulfate concentrations in general, with occasional long-range influence during ducting situations involving southwesterly flows of air. Similarly, warm frontal precipitation may involve cloud formation in air parcels advected for hundreds of kilometers from a southwesterly direction. The relative contributions of such long-range effects and of more local regional effects are currently unknown and cannot be reliably estimated using currently available models.

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5 Research Needs

In light of existing uncertainties in knowledge about the relationships between emissions and the deposition of acid-forming materials, it is appropriate to consider the research that may help to clarify understanding. In this chapter we describe research that we believe is needed to answer the most central questions in the shortest amount of time.

A considerable amount of research is currently being performed with funding from both governmental and private sources in the United States and Canada as well as in Europe. We have not attempted to review these research programs. As a consequence, some of the research activities that we recommend may be--indeed we know they are--included in current programs. Others, to the best of our knowledge, are not. On the basis of our review of current knowledge as described in this report, we are convinced that the research that we recommend forms essential elements in an integrated effort to improve understanding of the phenomenon of acid deposition.

We do not know whether incorporation into a model of all the chemical and physical processes currently thought to be important would account for the concentrations of sulfate observed in precipitation during both winter and summer. No quantitative determination of the relative contributions to the production of sulfate and nitrate by gas- and liquid-phase processes has been attempted to date; such attempts are probably several years in the future.

The details of dispersion of pollutants from sources, their chemical transformation and transport over long distances, and their contribution to acids in precipitation systems are sufficiently unknown and complex that a great deal of research will probably be required before

they can be described with precision by models. Furthermore, many years of precipitation measurements will be needed to establish a reliable data set from which tests of models can be made.

We believe that extensive laboratory, field, and modeling studies should be continued, to establish the physical and chemical mechanisms governing acid deposition. However, it appears to us that useful information about the delivery of acids to rural areas by transport and transformation processes can be determined fairly quickly by direct empirical observation in the field. Although the results of such field studies may not yield complete detailed descriptions of the interactions of all the processes involved, they are likely to provide basic phenomenological evidence with sufficient reliability to form the basis for improving the near-term strategy for dealing with the problem of acid deposition. The data are essential for enhancing theoretical understanding and developing improved deposition models. In the long term, the ultimate strategy for dealing with acid deposition will depend on the application of realistic, validated models.

FIELD STUDIES

Field research capable of testing the possibility of a nonlinear relationship between emissions and wet deposition in North America is needed. A limitation of oxidant in winter is believed to be a principal cause of such nonlinearity. Because the nature and availability of oxidants is sensitively dependent on the general composition of polluted air, field studies should be conducted in regions particularly involved in the problem of acid precipitation. Of primary importance are those experiments directed at discovering the rates of production of $SO_4^=$ and NO_3^- in clouds and the detection of possible limitations of those rates. Supplemental information, such as the availability of oxidants under varying conditions, is also important.

We describe below some of the more important issues currently amenable to study in the field.

Cloud Processes

As described in Chapter 2 and Appendix A, the conversion of SO_2 to H_2SO_4 in certain cloud systems may be

relatively complete and rapid, but we lack conclusive information on either oxidation rate constants or the completeness of conversion in acidified clouds typical of the eastern United States and Canada. Most of the mass of sulfuric acid already existing in aerosols in dry air is efficiently incorporated into cloud droplets as the air is cooled and the cloud forms. Models indicate that nitric acid vapor is also incorporated with high efficiency into cloud droplets. If, during the warm months, when the rate of wet deposition of acid is highest, complete in-cloud conversion of SO_2 and NO_2 occurs, and if the rainout of HNO_3 vapor and sulfate aerosol is highly efficient, then the assumption that the wet deposition of sulfuric and nitric acid is linearly related to ambient concentrations of SO_2 and NO_x would be justified. This could be readily checked experimentally with available technology in appropriate storm systems, such as warm fronts and summertime convective storms.

Studies of Chemical Mechanisms

A number of key problems relating to the linearity of the dependence of acid production rates on precursor concentrations provide the focus for studies of chemical mechanisms. (1) The efficiency and seasonal dependence of transformation processes are no doubt different for gas-phase and aqueous-phase processes. What are the relative contributions of these two pathways? (2) What are the lifetimes of NO_2 and SO_2 in clouds? Do rates of acid production differ markedly in various cloud types? Are the production rates inhibited by increasing acidity or limited by availability of oxidants? (3) What are the dominant reaction paths for SO_2 oxidation in clouds? (4) Do competing reactions with other atmospheric constituents (e.g., formaldehyde), especially in polluted air, seriously inhibit or lower the effectiveness of H_2O_2 reactions with SO_2 in clouds? (5) What processes govern the apparently significant rate of production of HNO_3 in clouds? (6) What are the relative rates of production of HNO_3 and H_2SO_4 in clouds? (7) What is the role of ultraviolet light in the production of free radicals and H_2O_2 in the gas phase in clouds?

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Dry Deposition

We noted in Chapter 3 that dry deposition of SO_2 and NO_2 may account for about one half of the total deposition of sulfur and nitrogen oxides and acids in eastern North America. Accurate evaluations of the extent of dry deposition of these pollutants is required to obtain a quantitative measure of their fates in the environment. The eddy-correlation technique, in which simultaneous measurements of the vertical wind speeds and specific pollutant concentrations are measured with rapid-response instrumentation, should prove to be a valuable tool in the study of the deposition of SO_2, NO_2, HNO_3, and other pollutant gases. The development, refinement, and application of this method and possibly other new and accurate methods of measuring dry deposition, which are suitable for monitoring applications, are important research objectives. The challenge of developing accurate methods is great, adapting as they must to the great variety of surfaces that cover the Earth and to the effects of humidity, temperature, sunlight intensity, and other factors.

Tracers

To develop the most cost-effective strategy for ameliorating the problem of acid deposition, it is necessary to know the relative impacts of specific source regions on specific sensitive receptor regions. In view of the uncertainties inherent in the calculation of trajectories, especially during storm conditions, it seems important to develop tracer techniques that can yield experimental tracking of air parcels. Such techniques could be applied especially during the meteorological conditions that are currently believed to provide the greatest opportunity for long-range transport of acidic materials. The use of insoluble and chemically inert gaseous tracers, as is currently planned, provides a promising approach. Admixtures of insoluble and unique materials that are subject to reaction with oxidants such as the HO radical would in addition provide information on possible limitations of the oxidation of SO_2 and NO_x.

In light of the potential advantages of the method of elemental tracers for understanding the relationships between source and receptor regions, a considerable

amount of research and development to establish and test the method on a regional scale is warranted. The first priority in this work should be to measure the detailed patterns of elemental composition of particulate matter collected at various rural sites in both source and receptor regions in eastern North America. The particles should be segregated by size, and a large number of elements and species should be measured for each size range. Other data should also be collected concurrently, including ambient concentrations of pollutant gases, wind speed and direction, relative humidity, precipitation, and optical qualities of the atmosphere. High-quality back trajectories should be calculated for the air masses sampled for each sampling period to aid in interpretation of the results. Few of these data now exist.

Research using elemental tracers would attempt to determine if particles coming from major source regions have distinctive chemical signatures that can be used to identify the origins of polluted air masses at long distances from source regions. For example, are there elemental tracers that can be used as clear indicators of coal combustion, in the same way as vanadium apparently can be used to indicate oil combustion? Can observed patterns of elemental composition in rural areas be resolved into linear combinations of known composition released by certain types of sources? Does the behavior of fine particles, particularly those bearing soluble species, provide insight into the behavior of sulfate and nitrate species that are of primary concern in acid deposition?

Building on the first stage of research, it may also be necessary to conduct studies of the elemental composition of particles in clear air and clouds as well as on the ground during specific episodes, such as the ducting of a polluted air mass from the midwestern United States toward the northeast along a southward-moving warm front. Changes in elemental composition as a specific parcel of air moves on a regional scale should help to identify the types of changes in the characteristics of the air during transport and as it passes over additional sources. Much of this work could be conducted in conjunction with the field studies described in the previous section.

Meteorological Studies

The climatology of storm movements in North America is well developed. The development of a quantitative relationship between storm type and acid deposition over eastern North America would help to estimate the long-range transport of pollutants. Despite the associated uncertainties, statistical studies of air parcel trajectories associated with the types of storm systems responsible for depositing acid precipitation would also be extremely valuable.

An important requirement in the experimental determination of rates of SO_2 and NO_x oxidation in clouds is a realistic evaluation of the airflow in the vicinity of clouds to establish the quantity of materials processed by and the time resident in the cloud.

All of these field studies bear directly on the critical question of the zone of influence of sources on receptor regions.

Our suggestion that these field measurements be a first priority is not meant to imply that pertinent laboratory and modeling studies should not also proceed. However, in view of the complexities in these systems, we believe that well-conceived field studies may answer many of the outstanding questions in a shorter time than that required for a complete molecular and dynamic description of the phenomenon.

LABORATORY STUDIES

The direct measurement of elementary rate constants for the many apparently important reactions related to the chemistry of acid deposition has been restricted largely to conditions that are not typical of the lower troposphere. The direct determination of the rate constants for reactions of the HO radical with SO_2 and NO_2 should be made at pressures (near 1 atm of air) and temperatures characteristic of the troposphere, as should other measurements.

Many aspects of cloud chemistry can and should be examined quantitatively under controlled laboratory conditions. For example, the mechanisms of the development of H_2O_2 from O_3 and other unidentified reactants should be established. Other important parameters amenable to laboratory measurement that are required for the quantitative description of cloud processes are the

sticking coefficients of the gas-phase reactants such as HO, HO_2, and NO_3 on collision with cloud droplets.

In general, laboratory studies are needed to establish the mechanistic detail that is required in the development of the chemically and physically sound models of acid precipitation.

DEVELOPMENT OF THEORETICAL MODELS

Models of the development, transport, and deposition of acids and acid-forming materials based on physical and chemical principles are under development. These models will serve as a useful framework with which the latest data from field and laboratory studies can be combined to provide a suitable test of theory and improved planning for further field studies. State-of-the-art theoretical models that treat quantitatively the complicated gas-phase chemistry, cloud processes, transport, and deposition will require many years for development. The ultimate test of our understanding of the chemistry and physics of the processes of acid rain is the successful development and use of such models.

Appendix A The Chemistry of Acid Formation

It is well established that the oxides of sulfur--sulfur dioxide (SO_2) and sulfur trioxide (SO_3)--and nitrogen--nitric oxide (NO) and nitrogen dioxide (NO_2)--are converted (oxidized) in the troposphere to sulfuric acid (H_2SO_4) and nitric acid (HNO_3), respectively. However, the most prevalent end products of these reactions--H_2SO_4, HNO_3, ammonium bisulfate (NH_4HSO_4), ammonium nitrate (NH_4NO_3), etc.--give few clues about which of several oxidizing pathways are important. Yet for the development of scientifically sound, predictive models of acid deposition that define theoretical source-receptor relationships, knowledge of the elementary chemical steps that are involved (among many other quantities such as deposition rates and transport rates) is required. The rates of these reactions follow well-defined mathematical laws (rate expressions) that relate reaction rates to concentrations of the reactants, temperature, pressure, and other variables. Considerable progress has been made in developing an understanding of the nature of this chemistry, but a large number of uncertainties still remain in key areas.

As our knowledge of the atmospheric chemistry of SO_2, NO, and NO_2 continues to grow, it has become increasingly clear that many different pathways exist for generating H_2SO_4 and HNO_3 in the troposphere. Reactions can occur in the gas phase; in the solution phase in cloud water and rainwater, for example; and in reactions on surfaces of solid particles in the atmosphere. Thus we are concerned with the rates of exchange of gaseous reactants and their reaction products between liquids and the surfaces of solids as well as the rates of interactions among gaseous molecules, aqueous phase molecules and ions, and species adsorbed on solid

surfaces. For current purposes, it is not necessary to review and evaluate every detail of these chemical processes, but we should note the nature of the many chemical processes, the variety of interactions among the many species involved, and the current state of knowledge related to the chemistry of acid deposition. We consider first the homogeneous gas-phase chemistry that results in oxidation of SO_2, NO, and NO_2 in the troposphere (Calvert and Stockwell 1983, Calvert et al. 1978).

GAS-PHASE REACTIONS LEADING TO GENERATION OF ACID IN THE TROPOSPHERE

Oxidation by Stable Atmospheric Molecules

The thermodynamic properties of the oxides of sulfur indicate that sulfur dioxide has a strong tendency to react with oxygen in the air under normal tropospheric conditions:

$$2SO_2 + O_2 \rightarrow 2SO_3. \qquad (1)$$

Thus the ratio of $[SO_3]/[SO_2]$ is about 8×10^{11} at equilibrium in air at 1 atm and 25°C. (Square brackets indicate the concentration of the species inside the brackets.) Thermodynamic arguments also tell us that at humidities normally encountered in the lower troposphere, the SO_3 produced by reaction (1) will be converted efficiently to sulfuric acid, $H_2SO_4(aq)$, according to

$$SO_3 + H_2O \rightarrow H_2SO_4(aq). \qquad (2)$$

The reaction of SO_3 with H_2O is so fast that any process in which SO_3 is formed in the moist troposphere can be considered equivalent to the formation of H_2SO_4. Certain metal ions (Mn^{2+}, Fe^{3+}, etc.) in aqueous solutions of SO_2 (HSO_3^-) can catalyze the overall sequence of reactions (1) and (2). However, thermodynamics tells us nothing about the rates of chemical reactions, and the rate of reaction (1) is so slow under catalyst-free conditions in the gas phase that it can be neglected as a source of sulfuric acid in the atmosphere.

The thermal oxidation of NO and NO_2 in the gas phase is also slow. Pathways that are thermodynamically favored are ,

$$2NO + O_2 \rightarrow 2NO_2, \quad (3)$$

$$2NO_2 + H_2O \rightarrow HNO_3 + HONO. \quad (4)$$

Reaction (3) requires literally days for the conversion of a significant fraction of the nitric oxide present [at concentrations of the order of parts per billion (ppb)] in the troposphere, and the homogeneous gas-phase reaction (4) is immeasurably slow (Schwartz and White 1982). Obviously, other, seemingly less direct reactions must be invoked to account for the observed rates of SO_2, NO, and NO_2 oxidation, which are between 1 and 100 percent/h.

A number of the more complex pathways involve photochemistry. In one, sulfur dioxide absorbs light in the ultraviolet region of the solar radiation incident in the troposphere, and, in principle, excited states of SO_2 generated in this fashion could lead to SO_2 oxidation in the troposphere. Figure A.1 indicates that there is a significant overlap between the actinic flux incident in the lower troposphere and two distinct absorption regions of SO_2. Excitation in the "forbidden," long-wavelength band forms the excited $SO_2(^3B_1)$ species, while excitation at the wavelengths below 330 nm generates higher excited states, presumably the $^1A_2$ and $^1B_1$ species.

$$SO_2(\tilde{X}^1A_1) + h\nu(340 < \lambda < 400 \text{ nm}) \rightarrow SO_2(^3B_1), \quad (5)$$

$$SO_2(\tilde{X}^1A_1) + h\nu(240 < \lambda < 330 \text{ nm}) \rightarrow SO_2(^1A_2, {}^1B_1). \quad (6)$$

These excited states of SO_2 are nondissociative; only quanta of light at wavelengths below 218 nm (which do not penetrate to the troposphere) provide sufficient energy to allow photodissociation:

$$SO_2(\tilde{X}^1A_1) + h\nu(\lambda < 218 \text{ nm}) \rightarrow SO_2(^1B_2) \rightarrow O(^3P) + SO(^3\Sigma^-). \quad (7)$$

The lower excited singlet states of $SO_2(^1A_1, {}^1B_1)$ appear to be very short lived in air at 1 atm, and they are rapidly converted by collisional perturbations to $SO_2(^3B_1)$ molecules, possibly $SO_2(^3A_2)$ and $SO_2(^3B_2)$ molecules, and ground-state $SO_2(\tilde{X}^1A_1)$ molecules. The rate of excitation of SO_2 through absorption of sunlight can be very significant. If this excitation were the rate-determining step in the photooxidation of SO_2, that is, if every molecule of SO_2 that is photoexcited were oxidized

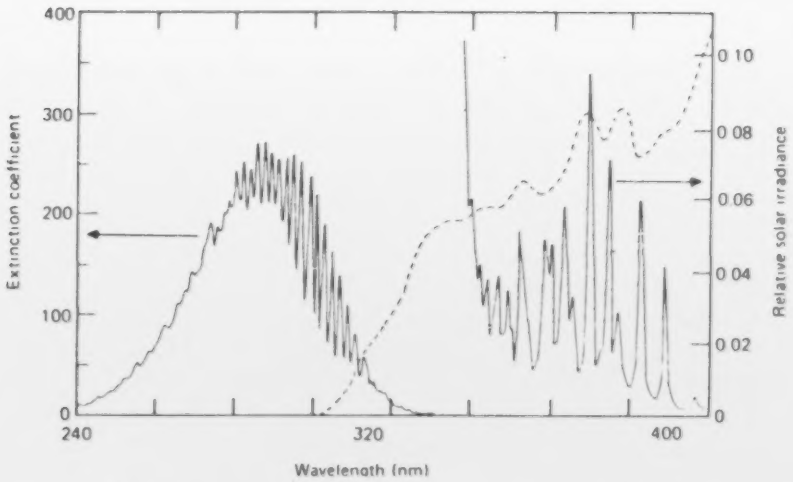

FIGURE A.1 Comparison of the extinction coefficients (liters mole^{-1} cm^{-1}, base 10) of SO_2 within the first allowed band (left), the "forbidden" band (right), and a typical distribution of the flux of solar quanta (relative) at ground level (dashed curve). SOURCE: Calvert et al. (1978).

through subsequent reaction with O_2 or other reactants, then the lifetime of SO_2 in the lower troposphere with overhead sun should be as low as 52 minutes (Sidebottom et al. 1972). Of course, this is not the case. The SO_2 ($^3B_1$) species appears to be one of the most favored states, which is ultimately populated through absorption of sunlight and collisional processes in the lower atmosphere. The reactions of this species with various atmospheric gases and many atmospheric impurities have been studied extensively (Calvert et al. 1978). Quenching of SO_2 ($^3B_1$) by atmospheric gases is expected to be the dominant process. In air at 1 atm, 25°C, and 50 percent relative humidity, quenching by N_2, O_2, H_2O, and Ar will occur 45.7, 41.7, 12.2, and 0.3 percent of the time, respectively. Quenching by impurity gases is highly improbable. Even when impurities are present at concentrations of the order of ppm, the rates of SO_2 conversion by such species are very slow (Calvert et al. 1978). All available evidence suggests that the only significant chemical result of the major quenching reactions of SO_2 ($^3B_1$) occurs with O_2, and this does not lead efficiently to any overall chemical change in the SO_2, but low-lying, excited electronic states of molecular oxygen, $O_2(^1\Delta_g)$ and $O_2(^1\Sigma_g^+)$, are formed by energy transfer.

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$$SO_2(^3B_1) + O_2(^3\Sigma_g^-) \rightarrow SO_2(\tilde{X}^1A_1) + O_2(^1\Sigma_g^+) \quad (8)$$

$$\rightarrow SO_2(\tilde{X}^1A_1) + O_2(^1\Delta_g). \quad (9)$$

These reactions are not unique sources of excited oxygen, species that are also formed in other atmospheric reactions at much higher rates (Calvert et al. 1978). Thus we conclude that photooxidation of SO_2 is also not an important source of acids in the atmosphere.

Similar conclusions are reached from a study of the photochemistry of NO and NO_2. NO does not absorb solar radiation in the wavelength range available near the Earth's surface, so its photochemistry is not important in the troposphere. NO_2 does absorb radiation over a wide range, and absorption of the wavelengths below 430 nm leads to dissociation ($NO_2 + h\nu \rightarrow O + NO$). However, formation of acid as a direct result of the photochemistry of NO and NO_2 is unimportant.

Reactive Transient Species in the Troposphere

Most of the gas-phase tropospheric chemistry of SO_2, NO, NO_2, and other impurity molecules involves reactions with a variety of reactive excited molecules, atoms, and free radicals (neutral fragments of stable molecules) formed by absorption of sunlight by trace gases in the atmosphere. As a background to discussions to come we review briefly here some of this important chemistry since it enters directly or indirectly into many of the reaction pathways that lead to the formation of acids in the troposphere.

In the polluted troposphere, NO_2 is dissociated by sunlight absorption ($\lambda < 430$ nm) to form reactive, ground state oxygen atoms, $O(^3P)$, and NO, while the oxygen atom reacts rapidly to form ozone (O_3):

$$NO_2 + h\nu(\lambda < 430 \text{ nm}) \rightarrow O(^3P) + NO, \quad (10)$$

$$O(^3P) + O_2(+M) \rightarrow O_3(+M). \quad (11)$$

Ozone can reoxidize NO to NO_2 in (12) or react with alkenes to give highly reactive ozonides in (13) and Criegee intermediates in (14):

$$O_3 + NO \rightarrow O_2 + NO_2 \qquad (12)$$

$$O_3 + RHC{=}CHR \rightarrow RHC(-O-O-O-)CHR \qquad (13)$$

$$RHC(-O-O-O-)CHR \rightarrow RCHO_2 + RCHO \qquad (14)$$

Ozone can also oxidize NO_2 to the reactive transient NO_3 in (15), and this can lead to N_2O_5 in (16):

$$O_3 + NO_2 \rightarrow O_2 + NO_3, \qquad (15)$$

$$NO_3 + NO_2 (+M) \rightleftharpoons N_2O_5 (+M). \qquad (16)$$

The photodecomposition of ozone may generate electronically excited oxygen atoms, $O(^1D)$, and excited molecular oxygen with absorption in the short-wavelength region of the spectrum:

$$O_3 + h\nu(290\text{-}306\ nm) \rightarrow O(^1D) + O_2(^1\Delta_g), \qquad (17)$$

$$O_3 + h\nu(290\text{-}350\ nm) \rightarrow O(^1D) + O_2 \text{ or } O + O_2(^1\Delta_g, {}^3\Sigma_g^-), \qquad (18)$$

$$O_3 + h\nu(450\text{-}700\ nm) \rightarrow O + O_2. \qquad (19)$$

The $O(^1D)$ species formed in (17) is much more reactive than the ground-state oxygen atoms [$O(^3P)$, often simply symbolized by O]. $O(^1D)$ reacts efficiently when it collides with a water molecule to form a highly important transient in atmospheric chemistry, the hydroxy radical, HO:

$$O(^1D) + H_2O \rightarrow 2HO. \qquad (20)$$

This radical, unlike many molecular fragments formed from carbon-containing molecules, is unreactive toward oxygen, and it survives to react with most atmospheric impurities such as the hydrocarbons, aldehydes, NO, NO_2, SO_2, and CO. Its reactions with carbon monoxide and the hydrocarbons (RH) lead to another important class of reactive transients, the peroxy radicals:

$$HO + CO \rightarrow H + CO_2, \qquad (21)$$

$$H + O_2(+M) \rightarrow HO_2(+M), \quad (22)$$

$$HO + RH \rightarrow H_2O + R, \quad (23)$$

$$R + O_2(+M) \rightarrow RO_2(+M). \quad (24)$$

Here R represents the alkyl groups such as methyl (CH_3), ethyl (C_2H_5), or another larger group derived from the parent hydrocarbons, methane (CH_4), ethane (C_2H_6), or larger hydrocarbon (RH), respectively. The reaction of the HO radical with aldehydes (RCHO) forms the acyl (RCO) and acylperoxy ($RCOO_2$) radicals in similar reactions:

$$RCHO + HO \rightarrow RCO + H_2O, \quad (25)$$

$$RCO + O_2(+M) \rightarrow RCOO_2(+M). \quad (26)$$

The peroxy radicals react rapidly with NO to form NO_2 and other classes of reactive species. In the case of the HO_2-NO reaction, HO is regenerated, while with the RO_2 and $RCOO_2$ radicals, alkoxy (RO) and acyloxy (RCO_2) radicals, respectively, are formed:

$$HO_2 + NO \rightarrow HO + NO_2, \quad (27)$$

$$RO_2 + NO \rightarrow RO + NO_2, \quad (28)$$

$$RCOO_2 + NO \rightarrow RCO_2 + NO_2. \quad (29)$$

The most common fate of the smaller alkoxy radicals in the lower atmosphere is reaction with oxygen, leading to HO_2 radicals and a carbonyl compound. For example, with the simplest alkoxy radical, methoxy (CH_3O), the following reaction occurs:

$$CH_3O + O_2 \rightarrow HO_2 + CH_2O. \quad (30)$$

The RCO_2 radicals are of short lifetime, decomposing to form an alkyl radical (R) and CO_2, with the subsequent generation of another peroxyalkyl radical:

$$RCO_2 \rightarrow R + CO_2, \quad (31)$$

$$R + O_2(+M) \rightarrow RO_2(+M). \quad (32)$$

Reactions (14)-(26) combined with reactions (27)-(32) form a chain reaction. That is, a single initial HO radical

may oxidize CO, hydrocarbon, or aldehyde, and additional HO radicals will be regenerated as NO is oxidized to NO_2; the subsequent steps occur again and again in a repeating cycle of events. The peroxy radicals and ozone are the principal oxidizing agents for NO in the lower atmosphere [reactions (15) and (27)-(29)] (Demerjian et al. 1974).

When peroxy radicals react with NO_2, an additional class of highly reactive compounds is generated--the peroxynitrates:

$$HO_2 + NO_2 \rightleftharpoons HO_2NO_2, \tag{33}$$

$$RO_2 + NO_2 \rightleftharpoons RO_2NO_2, \tag{34}$$

$$RCOO_2 + NO_2 \rightleftharpoons RCOO_2NO_2. \tag{35}$$

Peroxynitric acid formed in (33), and the alkyperoxynitrates formed in (34) are relatively unstable in the lower troposphere at temperatures common in summer months; they dissociate readily to reform peroxy radicals and NO_2. However, during the cold winter months or in the stratosphere they can act as temporary sinks for HO_2 and RO_2 radicals and NO_2. Peroxyacylnitrates ($RCOO_2NO_2$), of which peroxyacetylnitrate ($CH_3COO_2NO_2$) is the most common, have longer lifetimes and can be the source of radical generation even during the nighttime hours.

The excited singlet delta molecular oxygen, $O_2(^1\Delta_g)$, a product of reactions (17) and (18), and excited singlet sigma oxygen, $O_2(^1\Sigma_g^+)$, the product of reaction (8), can also be created by direct absorption of sunlight by atmospheric O_2 and by energy transfer reactions from other photoexcited species such as $NO_2(\lambda > 430$ nm) and excited triplet aromatic hydrocarbons.

For current purposes the complex array of interactions that occur among the reactive species outlined here and with the various atmospheric impurities need not be considered. It suffices to say that many aspects of tropospheric chemistry, including photochemical "smog" and ozone generation, depend on these happenings (Demerjian et al. 1974). It is important, however, to evaluate the potential significance of the many highly reactive transient species of the atmosphere for reactions that oxidize SO_2 and NO_2 to acids.

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Atmospheric Oxidation of SO_2 by Reactive Transient Species

There are a large number of potentially significant gas-phase reactions of the reactive transients leading to oxidation of SO_2 in the troposphere. The potential candidate reactions, summarized in Table A.1, have the thermodynamic potential to occur as measured qualitatively by the sign of the change in enthalpy ($\Delta H°$) for the overall reaction. Rate constants for most of these elementary reactions have been determined. These data, coupled with estimates of the concentrations of the transients in the atmosphere, allow us to evaluate the significance of each reactant in oxidizing SO_2 in the atmosphere. Such evaluations have shown that reactions (42), (50), (56), and (59) or (61) are potentially significant sources of SO_2 oxidation; some of these reactions are only important for certain peculiar atmospheric conditions. By far the most important of the gas-phase reactions is the reaction of HO radicals with SO_2:

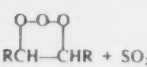
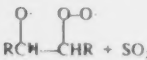
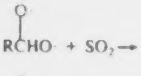
$$HO + SO_2(+M) \rightarrow HOSO_2(+M). \qquad (56)$$

The rate-constant data for reaction (56) have been reviewed recently (Calvert and Stockwell 1983) and are summarized in Figure A.2. The theoretical effect on this rate constant of the altered temperature and pressure of the atmosphere at various altitudes is shown in Figure A.3.

The "best" values of $k_{II(56)}$ suggested from the analysis given by Calvert and Stockwell (1983) are somewhat larger than those chosen by Moortgat and Junge (1977), Zellner (1978), and the values recommended for use by the CODATA (1980) group and the NASA (1979) panel and are more consistent with currently available information on the HO-SO_2 reaction. One must, however, retain considerable pessimism about the accuracy of all these data; realistic confidence limits should include ±50 percent of the suggested value.

Evidence is good that $HOSO_2$ formed in reaction (56) ultimately leads to the generation of sulfuric acid aerosol. However, $HOSO_2$ is not a stable molecule; it is a free radical that is probably highly reactive with several atmospheric compounds. It is not now clear what elementary reaction pathways are important in its conversion to H_2SO_4. Although there has been a great

TABLE A.1 Enthalpy Changes and Recommended Rate Constants for Potentially Important Reactions of Ground State SO_2 and SO_3 Molecules in the Lower Atmosphere

Reaction	$-\Delta H^{\circ}$ (kcal/mole) (25°C)	k^{b} (cm^3/molec-sec)
(36) $O_2(^1\Delta_g) + SO_2 \rightarrow SO_4$ (biradical, cyclic)	~25, ~28	
(37) $O_2(^1\Delta_g) + SO_2 \rightarrow SO_3 + O(^3P)$	~13.5	3.9×10^{-20}
(38) $O_2(^1\Delta_g) + SO_2 \rightarrow O_2(^1\Sigma_g^-) + SO_2$	22.5	
(39) $O_2(^1\Sigma_g^+) + SO_2 \rightarrow SO_4$ (biradical, cyclic)	~40, ~43	
(40) $O_2(^1\Sigma_g^+) + SO_2 \rightarrow SO_3 + O(^3P)$	1.5	6.6×10^{-16}
(41) $O_2(^1\Sigma_g^+) + SO_2 \rightarrow SO_2 + O_2(^1\Delta_g)$	15.0	
(42) $O(^3P) + SO_3 (+M) \rightarrow SO_3 (+M)$	83.0	5.7×10^{-14}
(43) $O_3 + SO_2 \rightarrow O_2 + SO_3$	57.6	$<8 \times 10^{-24}$
(44) $NO_2 + SO_2 \rightarrow NO + SO_3$	9.9	8.8×10^{-30}
(45) $NO_3 + SO_2 \rightarrow NO_2 + SO_3$	32.6	$<7 \times 10^{-21}$
(46) $ONOO + SO_2 \rightarrow NO_2 + SO_3$	~30	$<7 \times 10^{-21}$
(47) $N_2O_5 + SO_2 \rightarrow N_2O_4 + SO_3$	24.0	$<4 \times 10^{-23}$
(48) $HO_2 + SO_2 \rightarrow HO + SO_3$	16.7	$<1 \times 10^{-18}$
(49) $HO_2 + SO_2 (+M) \rightarrow HO_2SO_2 (+M)$	~7	
(50) $CH_3O_2 + SO_2 \rightarrow CH_3O + SO_3$	~27	$<1 \times 10^{-18}$
(51) $CH_3O_2 + SO_2 (+M) \rightarrow CH_3O_2SO_2 (+M)$	~31	$\sim 1.4 \times 10^{-14}$[c]
(52) $(CH_3)_3CO_2 + SO_2 \rightarrow (CH_3)_3CO + SO_3$	~26	$<7.3 \times 10^{-19}$
(53) $(CH_3)_3CO_2 + SO_2 \rightarrow (CH_3)_3CO_2SO_2$	~30	
(54) $CH_3COO_2 + SO_2 \rightarrow CH_3CO_2 + SO_3$	~33	$<7 \times 10^{-19}$
(55) $CH_3COO_2 + SO_2 \rightarrow CH_3COO_2SO_2$	~37	
(56) $HO + SO_2 (+M) \rightarrow HOSO_2 (+M)$	~37	1.1×10^{-12}
(57) $CH_3O + SO_2 (+M) \rightarrow CH_3OSO_2 (+M)$	~24	$\sim 5.5 \times 10^{-13}$
(58) 	~69	See text
	~89	See text
(59) $RCHOO\cdot + SO_2 \rightarrow RCHO + SO_3$	~79	$k_{59}/k_{60} \sim 6 \times 10^{-5}$
(60) $RCHOO\cdot + H_2O \rightarrow RCOOH + H_2O$	~121	(R = CH_3)
(61) 	~58	$k_{61}/k_{62} \simeq 4$
(62)	~12	(R = H)
(63) $SO_3 + H_2O \rightarrow H_2SO_4$	24.8	9.1×10^{-13}

[a] Enthalpy change estimates were derived from the data of Benson (1978), Harding and Goddard (1978), and Domalski (1971).

[b] Rate constants are expressed as second-order reactions for 1 atm of air at 25°C; see Calvert and Stockwell (1983) for the references to the original literature.

[c] The reverse reaction is so fast that the rate of oxidation of SO_2 via (51) is very dependent on alternate fates of the $CH_3O_2SO_2$ species.

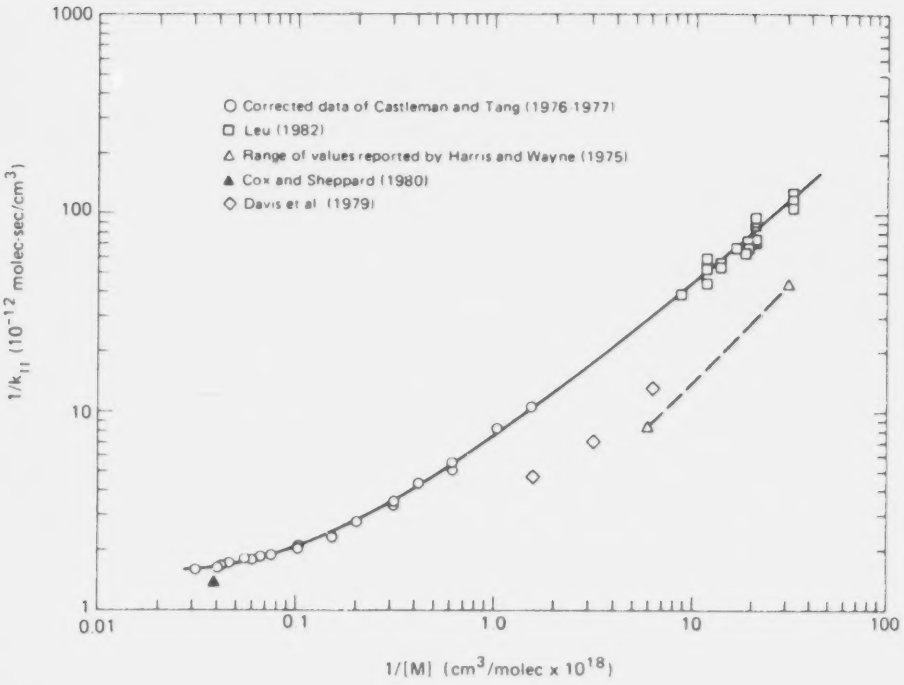


FIGURE A.2 Comparison of the experimental data for the effective second-order rate constants for the reaction (56) with M = N_2. SOURCE: Calvert and Stockwell (1983).

amount of speculation in this regard, there is little experimental evidence to help determine the relative importance of suggested alternative routes. Thus Cox (1974-1975, 1975), Calvert and McQuigg (1975), Calvert et al. (1978), Davis et al. (1979), Friend et al. (1980), Leu (1982), Benson (1978), and others have suggested that the $HOSO_2$ radical may participate in a variety of radical-radical and radical-molecule reactions. These reactions are summarized in Figure A.4. Although the mechanism of H_2SO_4 generation following (56) is unclear today, it is probable that reaction (56) is the rate-determining step in the sequence. Recent evidence suggests that the concentration of HO in photooxidizing mixtures of HONO, NO, NO_2, and CO is insensitive to even large additions of SO_2 (Stockwell and Calvert 1983). Thus the following sequence of reactions seems favored:

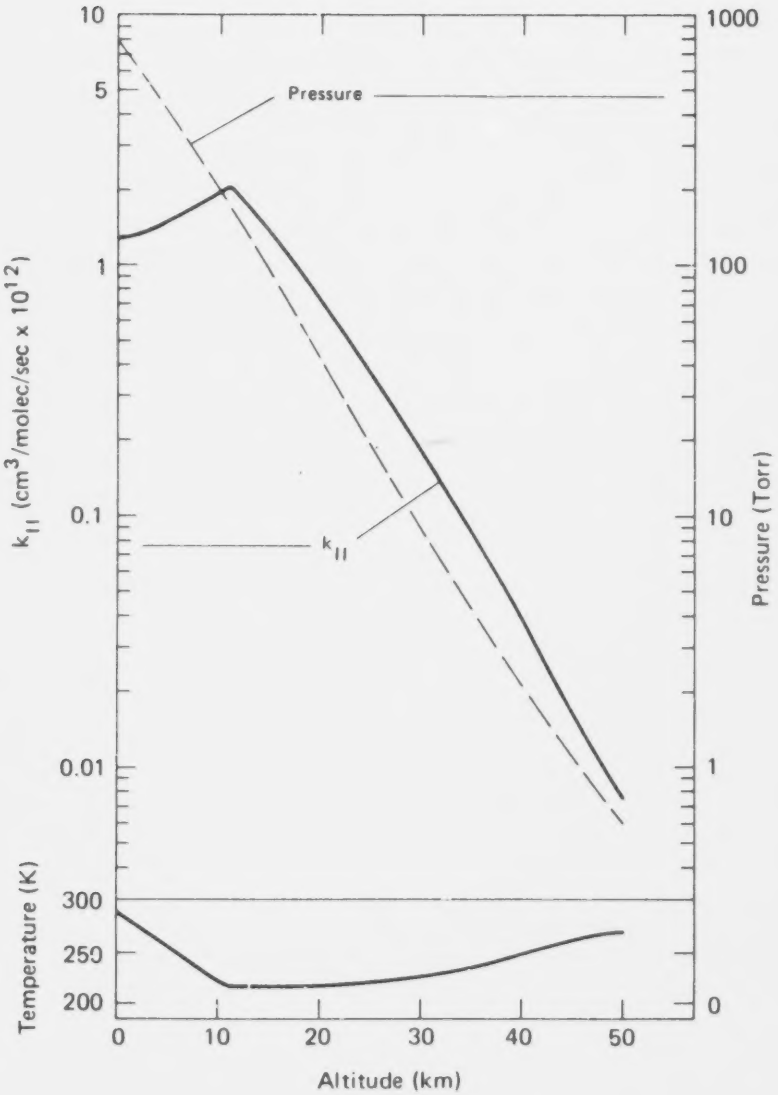


FIGURE A.3 Pressure, temperature, and the apparent second-order rate constant for the reaction (56) as a function of altitude. SOURCE. Adapted from Calvert and Stockwell (1983) for the conditions of pressure and temperature defined for the standard atmosphere (Valley 1965).

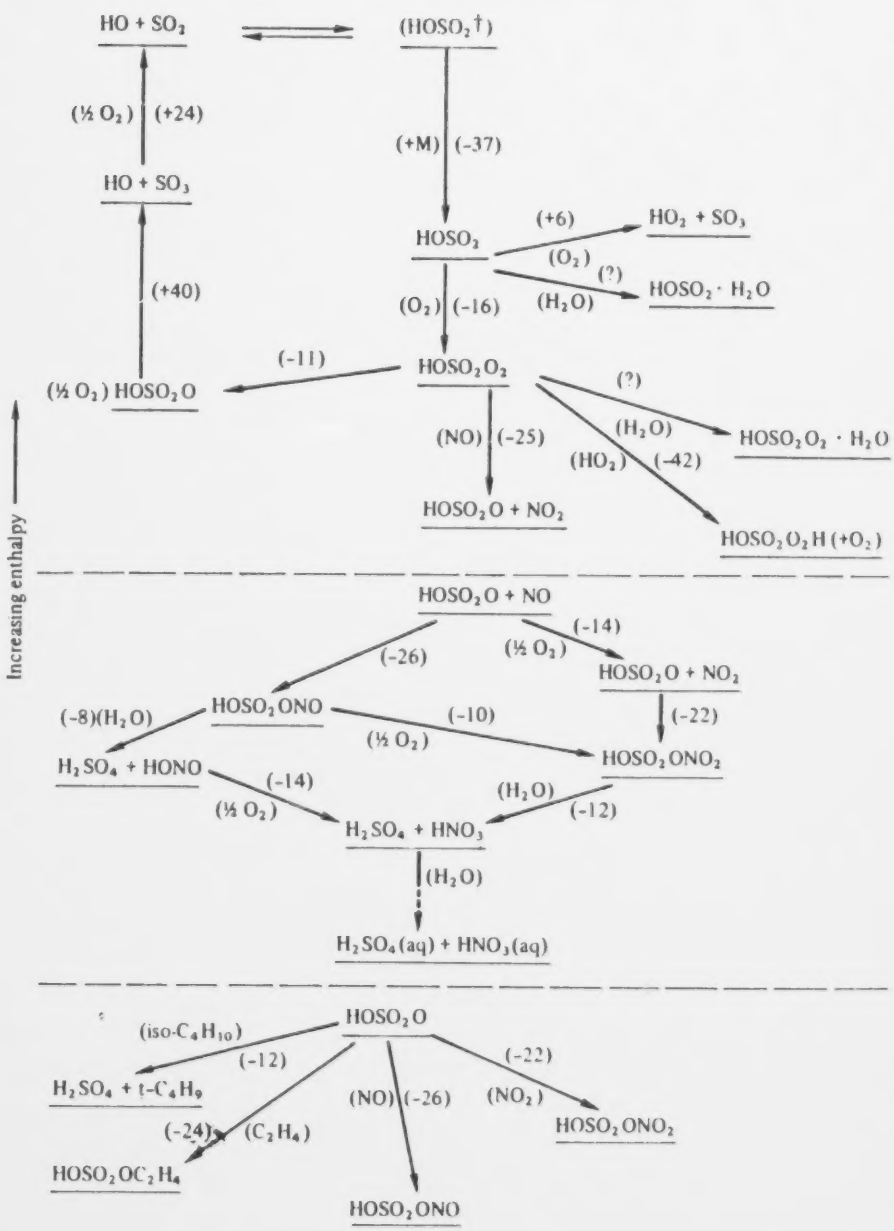


FIGURE A.4 Enthalpy relationships between various possible products of the HO-radical reaction with SO_2. SOURCE: Calvert and Stockwell (1983).

$HOSO_2 + O_2 \rightarrow HO_2 + SO_3$,
$HO_2 + NO \rightarrow HO + NO_2$,
$SO_3 + H_2O \rightarrow H_2SO_4$.

Atmospheric Oxidation of NO_2 by Reactive Transient Species

A great variety of experimental evidence supports the view that a very important pathway for the tropospheric oxidation of NO_2 is similar to reaction (56) for SO_2:

$$HO + NO_2(+M) \rightarrow HONO_2(+M). \qquad (64)$$

Estimates of $k_{II(64)}$ are well characterized for conditions applicable to the troposphere ($M = N_2$). Using Anderson's (1980) recommendations for this rate constant, we have made the plot shown in Figure A.5 of the effective second-order rate constant for various altitudes and conditions of [M] and temperature characteristics of the standard atmosphere. The same general trends in $k_{II(64)}$ with altitude are seen as observed for the HO-SO_2 reaction rate constant $k_{II(56)}$ in Figure A.3, but the values of $k_{II(56)}$ are roughly a factor of 10 larger than those for $k_{II(64)}$ for a given temperature and pressure in the troposphere.

A second homogeneous mode of potential formation of $HONO_2$ involves N_2O_5:

$$H_2O + N_2O_5 \rightarrow 2HONO_2. \qquad (65)$$

It is difficult to distinguish the homogeneous component of this reaction in laboratory experiments, since reaction at a moist cell wall can be more important than the homogeneous reaction for most experimental systems. Morris and Niki (1973) reported an upper limit for this constant of 1.3×10^{-20} cm^3 $molec^{-1}$ sec^{-1}. For theoretically expected concentrations of N_2O_5 in the troposphere ($\leq 2.5 \times 10^9$ molec cm^{-3}) during the daylight hours (Demerjian et al. 1974), the rate of (65) is insignificant compared with that of (64). In theory the reaction (65) can be important at night when NO_3 and N_2O_5 concentrations may rise.

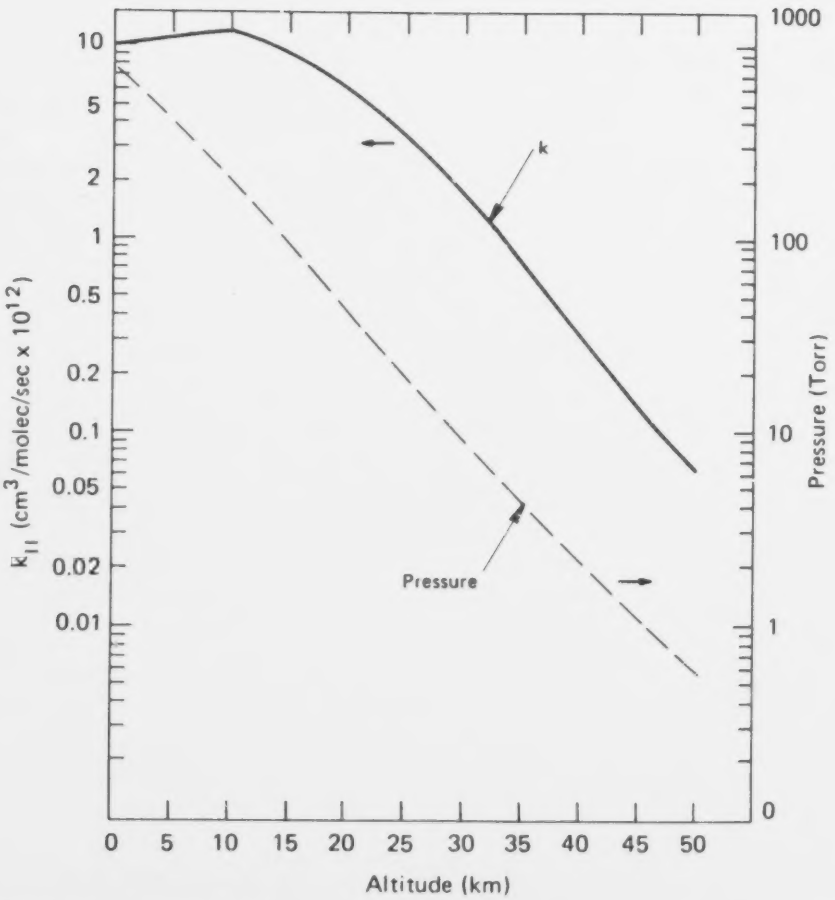


FIGURE A.5 Pressure and the apparent second-order rate constant for reaction (64) as a function of altitude. SOURCE: Calculated from the equations of Anderson (1980) for the conditions of pressure and temperature defined for the standard atmosphere (Valley 1965) (from Calvert and Stockwell 1983).

Theoretical Rates of SO_2 and NO_2 Conversion to H_2SO_4 and $HONO_2$ Through Gas-Phase Reactions in the Troposphere

The major homogeneous processes for SO_2 and NO_2 conversion to H_2SO_4 and $HONO_2$, respectively, are governed by the rates of reactions (56) and (64), which depend on the concentrations of the reactants, NO_2, SO_2, and HO. Knowledge of [HO] in the troposphere is needed to make theoretical estimates of the minimum rates of the gas-phase generation of the H_2SO_4 and $HONO_2$.

Although the direct measurement of [HO] in the troposphere is very difficult, there are both theoretical and experimental estimates of this quantity that are of some value for our considerations here (Table A.2). We have excluded early estimates based on laser-induced fluorescence of the HO radical because the generation of HO by the probing laser beam itself was then an unrecognized problem. Data based on the ^{14}CO-chemical method of Campbell et al. (1979), the laser-induced fluorescence data of Wang et al. (1981), and the HO absorption data of Perner et al. (1976) are all reasonably consistent both among themselves and with theoretical estimates based on computer models of the complex atmospheric chemistry (Calvert and McQuigg 1975, Chang et al. 1977, Crutzen and Fishman 1977, Demerjian et al. 1974, Graedal et al. 1976, Hecht and Seinfeld 1972, Höv and Isaksen 1979, Levy 1974, Niki et al. 1972).

We may use these data to make reasonable estimates of the gas-phase oxidation rates for SO_2 and NO_2 in order to judge the potential of reactions (56) and (64) to develop the ingredients for "acid rain." At high concentrations of HO radical characteristic of midday sunny summer skies in a polluted atmosphere (about 9×10^6 molec cm^{-3}), we anticipate from our selected rate constant data that SO_2 will oxidize at a rate of $\approx 3.7 \pm 1.9$ percent/h through HO-radical attack in reaction (56). Rates of $HONO_2$ generation in (64) are expected to be about 34 ± 17 percent/h for these conditions. Somewhat lower midday solar intensities typical of the winter months in a polluted atmosphere for which the maximum noontime value for [HO] $\approx 2.4 \times 10^6$ molec cm^{-3} lead to rates of about 1 ± 0.5 percent/h for SO_2 and about 18 ± 9 percent/h for NO_2. Of course, these rates are expected to track [HO] during the day and thus to drop rapidly to near zero at night.

Computer simulations (Stockwell and Calvert 1983) show that for conditions that provide a maximum summertime value for [HO] of about 9.1×10^6 molec cm^{-3}, the 24-h average [HO] is about 1.7×10^6 molec cm^{-3}, leading to a daily average rate of SO_2 oxidation through reaction (56) of 0.7 percent/h or 16.4 percent/24-h period. The equivalent NO_2 oxidation rate in (64) is 6.2 percent/h or 150 percent/24-h period. The average wintertime rate of oxidation is much lower: for SO_2 it is about 0.12 percent/h or 3 percent/24-h period and for NO_2 about 1.1 percent/h or 25 percent/24-h period. In theory therefore we expect the homogeneous gas-phase conversion of SO_2

TABLE A.2 Measured and Theoretical Estimates of the [HO] in the Troposphere

	[HO], molec/cm^3
A. Experimental Methods	
1. Chemical Method: $^{14}CO + HO \rightarrow {}^{14}CO_2 + H$ (Campbell et al. 1979)	
Pullman Wash. (46.7° N, 770 m)	$(5.8 \pm 2.4) \times 10^6$
New Zealand (44° S, 1030 m)	$(1.1 \pm 0.4) \times 10^6$
Tennessee (rural, 36° N, 270 m)	$(1.9 \pm 0.7) \times 10^6$
Los Angeles (34° N, 270 m)	$(5.0 \pm 1.2) \times 10^6$
Arizona (desert, 37° N, 2300 m)	$(57 \pm 23) \times 10^6$
2. Laser-Induced Fluorescence, 282.07-nm excitation, 309-nm emission, 302-nm N_2 Raman (Wang et al. 1981)	
Niwot Ridge, Colo. (3048 m)	$(40 \text{ to } 6) \times 10^6$
Los Angeles, Calif. (11,886 m)	$(2.5 \pm 2) \times 10^6$
San Bernadino, Calif. (10,057 m)	$(20 \pm 6) \times 10^6$
San Diego, South, Calif. (10,667 m)	$(10 \pm 4) \times 10^6$
Denver, Colo. (10,057 m)	$(1 \pm 3) \times 10^6$
3. Absorption Spectroscopy, 307.995 nm (Perner et al. 1976)	
Jülich, Germany (51° N)	$(11 \pm 6) \times 10^6$ usually $(6 \pm 3) \times 10^6$
B. Theoretical Estimates: Typical recent estimates found in simulations of Calvert, Demerjian, Seinfeld, Niki, Graedel, Isaksen, etc., and their co-workers (values are sensitive to ultraviolet solar irradiance at point of interest, levels of impurities, NO, NO_2, RH's, RCHO's, etc.):	
Daytime maximum (summer, 40° N)	$\sim (9 \pm 4) \times 10^6$
Daytime maximum (winter, 40° N)	$\sim (2 \pm 1) \times 10^6$
Nighttime	$\lesssim 2 \times 10^5$

SOURCE: Calvert and Stockwell (1983).

and NO_2 to provide a significant quantity of sulfuric and nitric acids in the troposphere.

When making such estimates for the gas-phase SO_2 oxidation rates, we must remember that other as yet unevaluated contributions from the alkene–O_3 products, the CH_3O_2-radical, and probably other unidentified reactants may contribute to the total SO_2 oxidation rate as well. For example, the reaction (42) of $O(^3P)$ with SO_2 can be significant in a highly NO_2-polluted atmosphere such as that present in the early stages of dilution of a stack gas plume. In this case, reaction (42) can theoretically account for an initial burst of SO_3 (H_2SO_4) formation at a maximum rate of about 1.4 percent/h (Calvert et al. 1978). In addition, the methylperoxy (CH_3O_2) and possibly other primary alkylperoxy

radicals may lead to SO_2 oxidation in the troposphere under special circumstances of high NO-NO_2 pollution, although the evidence for the significance of reaction (50) is not unambiguous (Kan et al. 1981). Also the products of the ozone-alkene reactions oxidize SO_2 readily, presumably through the Criegee intermediates in reactions (59) or (61). The aldehydes, water vapor, carbon monoxide, NO, and possibly other atmospheric impurities compete with SO_2 for these reactive intermediates, and the effectiveness of the competition of SO_2 with NO remains unclear today (Calvert and Stockwell 1983). If the rate constants for these $RCHO_2$ species with NO and SO_2 are similar and/or the ratio of $[NO]/[SO_2]$ is low, then SO_2 oxidation by Criegee intermediates can be significant in alkene-ozone-containing atmospheres (a few tenths of a percent per hour).

Although the contribution of SO_2 oxidation from reactions other than (56) may be important, the theoretical rates given here for the HO reactions alone appear to match reasonably well those observed in relatively dry SO_2 urban plumes (Eatough et al. 1981, Forrest et al. 1981, Garber et al. 1981, Gillani and Kohli 1981, Hegg and Hobbs 1980, McMurry and Rader 1981, Meagher et al. 1981, Newman 1981, Williams et al. 1981, Wilson and McMurry 1981, Zak 1981). Certainly a large part of the observed oxidation in the cloud-free ambient troposphere during the daylight hours arises from the HO-SO_2 reaction. The conversion rates for NO_2 observed by recent workers are also consistent with the estimates presented here (Spicer 1982).

THE SOLUTION-PHASE OXIDATION OF SO_2 IN THE TROPOSPHERE

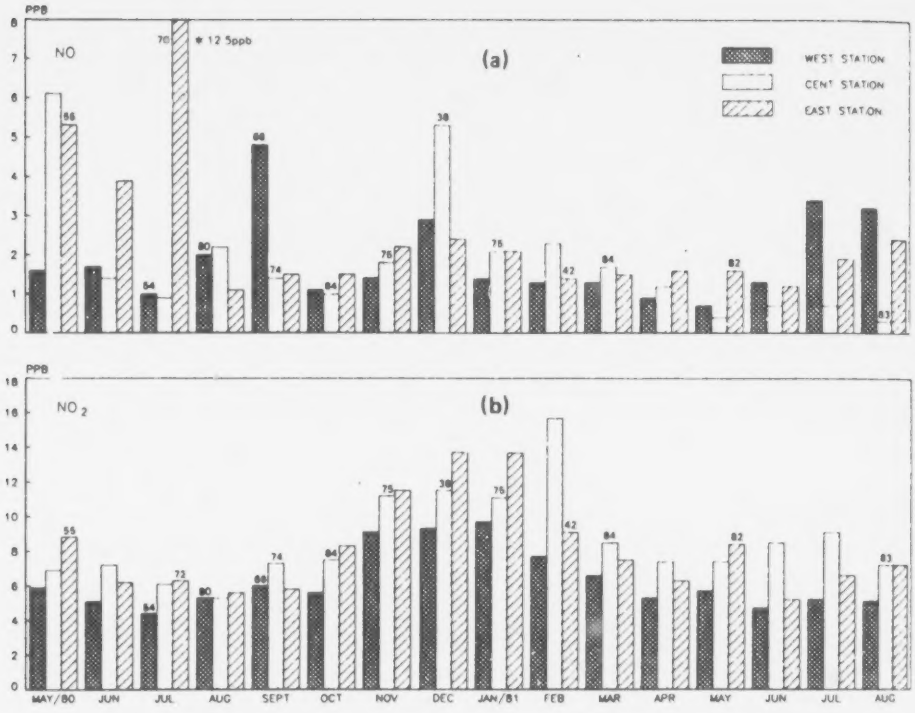
In recent years the role of the aqueous-phase reactions in the development of acid precipitation has received increased attention. The results of field and laboratory studies suggest that the relative importance of the gas-phase and solution-phase pathways may vary depending on a variety of meteorological conditions such as the extent of cloud cover, relative humidity, the amount of precipitation, the intensity of the solar radiation, and the presence and concentration of various pollutants. Rates of oxidation of SO_2 through gas-phase reactions are relatively slow (a few percent per hour during daylight), whereas in theory those for the solution-phase pathways may be as high as 100 percent/h for seemingly realistic concentrations of the reactants in cloud

water. Despite the considerable difference in rates of oxidation pathways in the aqueous and gaseous phases, both must be regarded as participating in the development of acid deposition in the eastern United States because air masses in this region are more likely to be free of clouds and precipitation a large fraction of the time (Niemann 1982). There appears to be little question that both gas-phase and liquid-phase processes can contribute significantly to the formation of H_2SO_4 in the atmosphere. Clear evidence of the relative importance of the two processes for various conditions is not now available.

Field data taken in dilute stack plumes in relatively cloud-free atmospheres show that formation of sulfuric acid at night is very slow and that during daylight hours the rate correlates with solar intensity (Hegg and Hobbs 1980, Wilson and McMurry 1981). Measurements are not frequently made at night, however, and there is no clear test, of which we are aware, of cloud chemistry for conditions that minimize the possible gas-phase processes. Wilson and McMurry (1981) have observed the evolution of aerosol size distributions as a result of gas-to-particle conversion. From these data they suggest that droplet-phase conversion of SO_2 to sulfate may be important at high humidities (>50 percent), where up to 20 percent of the growth of the aerosol was attributed to this reaction. Aerosol from gas-phase processes dominated the plume chemistry for low humidities and in the absence of clouds.

The clearest evidence of the major input from precipitating clouds was derived in the Acid Precipitation Experiment (APEX) (Lazrus et al. 1983). The investigators found significant production of both sulfuric and nitric acids in clouds. The acidity of dry air south of a warm front was measured before it ascended and produced a large area of warm frontal precipitation. Comparison of the chemical composition of the dry air and precipitation at the base of the cloud showed that rapid production of acid had occurred in the cloud. Rates of HNO_3 and H_2SO_4 production in the cloud were estimated to be of the order of 0.5 ppb/h and 1.2 ppb/h, respectively. Although analyses of H_2O_2 in the cloud suggested the involvement of this oxidizing agent, unidentified interferences in the luminol method for H_2O_2 detection discovered later leave the nature of the oxidizing agent unresolved.

Recently, transport and transformation chemistry of SO_2 were studied in an air mass tagged with SF_6 along the coast of southern California (Cass and Shair 1983). These studies appear to provide the first unambiguous evidence



that SO_2 conversion to sulfate can occur at a measurable rate at night (about 10 percent/h) in low stratus clouds over the coastal waters.

Strong seasonal variations in the sulfate and SO_2 in precipitation (measured as H^+ and HSO_3^-) have been seen in the MAP3S precipitation chemistry data (Henderson and Wingartner 1980). A more direct and quantitative measure of seasonal variations in the atmospheric chemistry involved in acid deposition has been presented by Shaw and Paur (1983). They measured gaseous NO, NO_2, SO_2, and airborne sulfate aerosol at three sites in the Ohio River Valley during the period May 1980 to August 1981 (Figure A.6). Two sites (labeled west and central in the figure) were located in rural farming communities (Union County, Kentucky, and Franklin County, Indiana), and the third (east) was in a forest clearing (Ashland County, Ohio). Figure A.6 indicates that while monthly average concentrations of NO remained relatively constant throughout the period of the experiment, NO_2 and SO_2 (gas-phase sulfur) increased in the winter months. Airborne particulate sulfate shows a minimum in winter, as does the percentage of total sulfur existing as

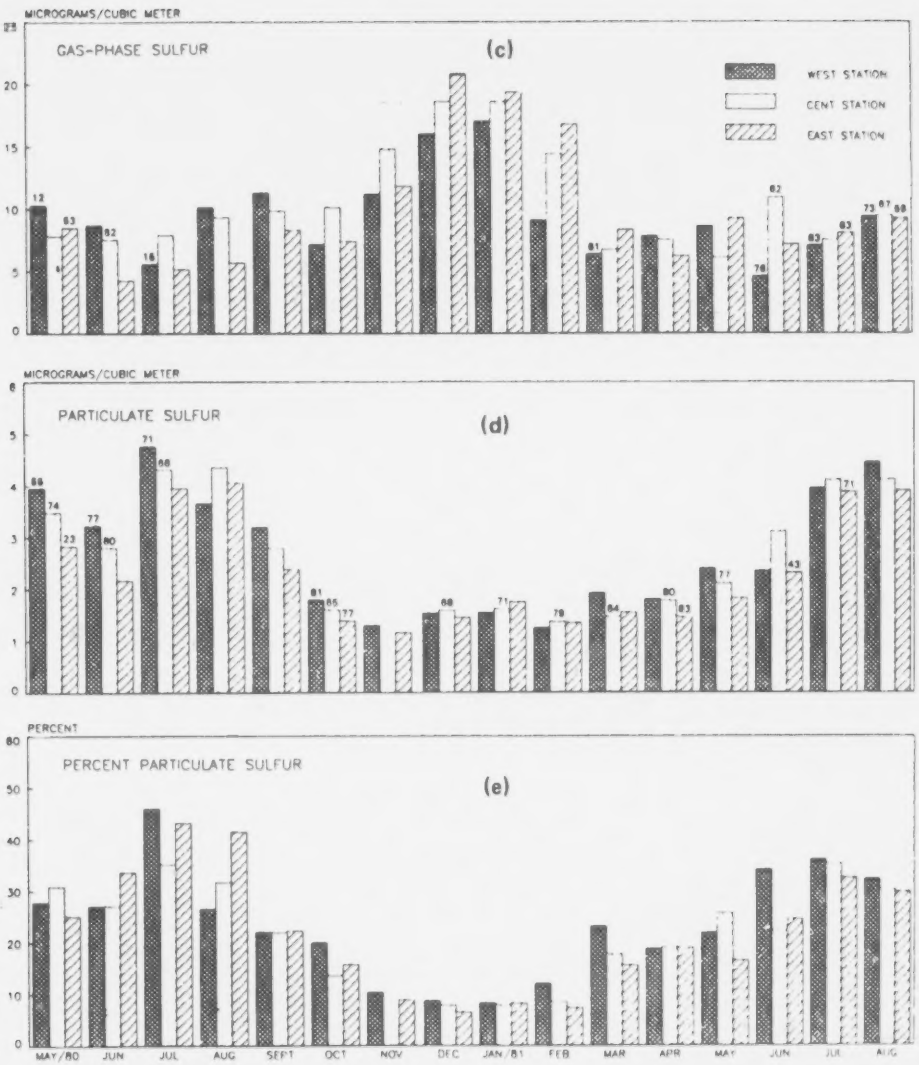


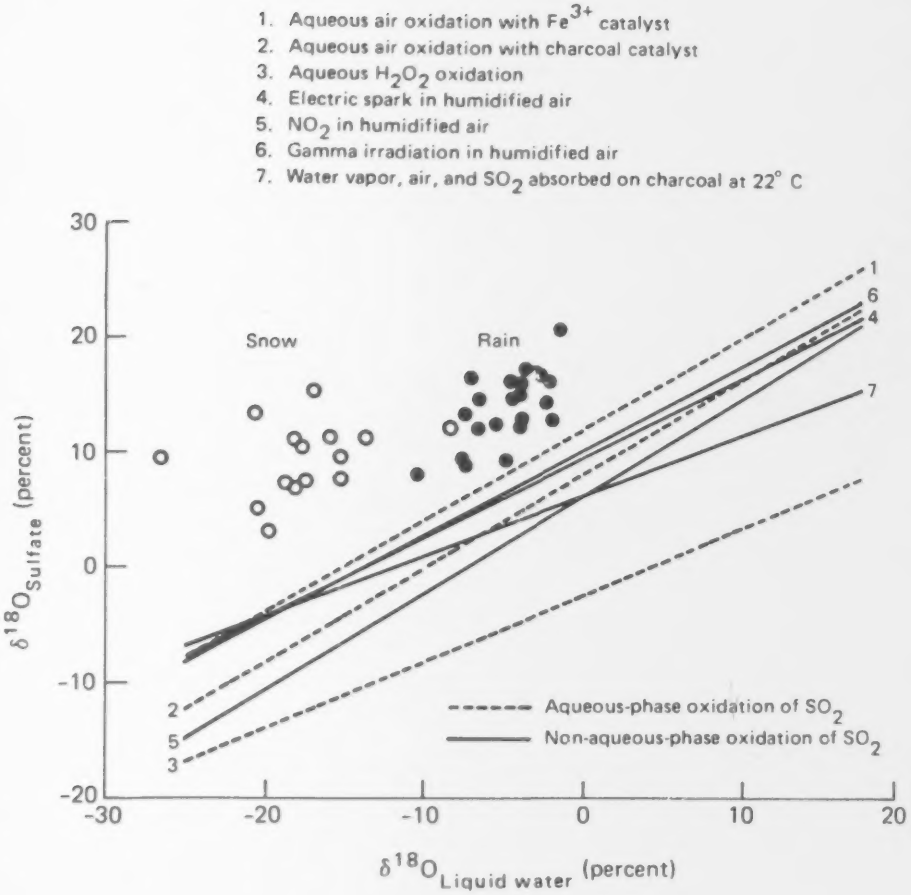
FIGURE A.6 Average monthly concentrations of airborne pollutants at three sites in the Ohio River Valley in 1980-1981. (a) NO, (b) NO_2, (c) SO_2, (d) sulfate aerosol, (e) percentage of total airborne sulfur existing as sulfate aerosol. SOURCE: Shaw and Paur (1983).

sulfate aerosol. The results are consistent with the general expectations of pathways for oxidation in both the gaseous and the aqueous phases since higher concentrations of the oxidizing agents (HO, H_2O_2, and O_3) are anticipated in the summer months when both solar intensities and the length of daylight are higher.

Evidence concerning the relative contributions to SO_2 oxidation by gaseous- and liquid-phase processes has also been presented using the isotopic abundance of ^{18}O in sulfate formed in various laboratory experiments and in natural samples of snow and rain collected near Argonne, Illinois (Holt et al. 1981, 1982, 1983). The method is based on the fact that oxygen atoms in sulfate ions in water do not reach equilibrium with those of water for temperatures and times characteristic of acidification of droplets in acid precipitation. Differences also exist in the ratios of ^{18}O to ^{16}O in O_2 and H_2O. Therefore, in principle, measured differences in the ^{18}O content of cloud-water sulfate should provide evidence of the origins of the sulfate. An isotopic ratio typical of H_2O would suggest solution-phase origins, whereas a ratio typical of atmospheric O_2 or species derived from the O_2 (O_3, H_2O_2, HO, etc.) would suggest that gaseous-phase chemistry was more important.

In their 1981 report, Holt et al. (1981) suggested that solution-phase oxidation of SO_2 is the more effective pathway for observed isotopic ratios in sulfate in natural precipitation water. However, the most recent interpretation of Holt et al. (1982, 1983) of the measured isotopic ratios in samples of atmospheric water and controlled laboratory experiments using a variety of homogeneous gas- and liquid-phase and catalytic methods of SO_2 oxidation is more ambiguous (Figure A.7). Note that the oxidation reaction of SO_2 with aqueous H_2O_2, currently the most favored reaction, gives isotopic ratios that are least in accord with the values observed for sulfate in snow and rain in field studies. The authors now conclude that some currently unknown source of sulfate (possibly primary sulfate) with enriched isotopic ratio must exist that dilutes that formed by one or more of the currently known reactions leading to sulfate formation.

An abundance of data based largely on laboratory measurements points to the probable involvement of the oxidizing agents hydrogen peroxide and/or ozone in the formation of acids in cloud water. Also suggested in theory as having a possible influence in these transformations are free radicals such as HO and HO_2 either formed in the gas phase and transported to the liquid phase or formed in the liquid phase. Soot (carbonaceous



$\delta^{18}O = [(f/f_{smow}) - 1] \times 10^3$, where $f = {}^{18}O/{}^{16}O$ and $f_{smow} = 2.005 \times 10^{-3}$, the isotopic ratio of standard mean oceanic water.

FIGURE A.7 Comparison of isotopic ratios in sulfates formed by seven laboratory methods and in precipitation water. SOURCE: Holt et al. (1982, 1983).

material) and various metal ions (copper, iron, manganese, and vanadium, for example) can act as catalysts for solution-phase oxidation of SO_2. Ammonia and other basic materials (such as metal carbonates) can influence the rates of solution-phase oxidation of SO_2 through alteration of the pH of the solution. We will review

briefly here the kinetic results that define the reaction rates for the solution-phase oxidation of SO_2. Many of the rate data presented here are from the review of sulfite oxidation by Martin (1983).

Aqueous-Phase Oxidation of SO_2

Sulfur dioxide dissolves in water to form the hydrate $SO_2 \cdot H_2O$ and the ions HSO_3^-, H^+, and $SO_3^{=}$; these species are related by the following equilibria, which are established rapidly:

$$SO_2(g) + H_2O(liq) \rightleftharpoons SO_2 \cdot H_2O(aq), \quad K_H = 1.23 \text{ M/atm } (25°C),$$

$$SO_2 \cdot H_2O(aq) \rightleftharpoons H^+(aq) + HSO_3^-(aq), \quad K_1 = 1.7 \times 10^{-2} \text{ M},$$

$$HSO_3^-(aq) \rightleftharpoons H^+(aq) + SO_3^{=}(aq), \quad K_2 = 6.0 \times 10^{-8} \text{ M}.$$

The concentration of undissociated H_2SO_3 is insignificant. The solubility of gaseous SO_2 or the concentration of total dissolved S(IV) equilibrated in water at some specified pH with gaseous SO_2 at a pressure P_{SO_2} is given by the relation:

$$[S(IV)] = P_{SO_2} K_H (1 + K_1/[H^+] + K_1K_2/[H^+]^2),$$

where $[S(IV)] = [SO_2 \cdot H_2O(aq)] + [HSO_3^-(aq)] + [SO_3^{=}(aq)]$. It is apparent that SO_2 becomes less soluble as the acidity of a solution increases. Freiberg and Schwartz (1981) and Schwartz and Freiberg (1981) have shown that the times required to establish gas-liquid equilibrium between SO_2 and aqueous aerosol droplets are usually much shorter than the chemical reaction times for reactant concentrations typical of the ambient atmosphere. The expected [S(IV)] in aqueous aerosols of pH 3.0 in equilibrium with SO_2 in the atmosphere ranges from about

10^{-9} M for 0.2 ppb SO_2 to about 10^{-6} M for 200 ppb SO_2. At pH 4.0, [S(IV)] varies from about 10^{-8} M for 0.2 ppb to about 10^{-5} M for 200 ppb SO_2. At pH 5.0, [S(IV)] is about 10^{-7} M for 0.2 ppb SO_2 and 10^{-4} M for 200 ppb SO_2.

Of the various oxidizing agents that can oxidize S(IV) in solution, two species, H_2O_2 and O_3, appear to be especially significant for ambient levels of impurities. The possibility that H_2O_2 and O_3 could be important oxidants was first suggested by Penkett et al. (1979). The overall reactions are represented by

$$H_2O_2(aq) + HSO_3^-(aq) \rightarrow SO_4^=(aq) + H^+(aq) + H_2O(aq), \quad (66)$$

$$O_3(aq) + HSO_3^-(aq) \rightarrow SO_4^=(aq) + H^+(aq) + O_2(aq). \quad (67)$$

Martin and Damschen (1981) have summarized all the pertinent rate data for reaction (66) (Figure A.8) and derived the following rate equation for the reaction:

$$\frac{d[S(IV)]}{dt} = \frac{8 \times 10^4 [H_2O_2][SO_2 \cdot H_2O(aq)]}{0.1 + [H^+(aq)]} \quad (68)$$

mole liter^{-1} sec^{-1}.

When $[H^+] \ll 0.1$ M, the expression (68) shows the oxidation rate to be independent of pH. This system is unique in this regard among the potential oxidizers of SO_2 in solution, and hence the H_2O_2 oxidation mechanism is favored at low values of pH common in atmospheric aerosols and precipitation water. The apparent activation energy for the reaction is about 8 kcal/mole in the pH range 4-6 and 6.7 kcal/mole at pH 2 (no buffer).

Hoffman and Edwards (1975) proposed a mechanism consistent with rate law (68):

$$HSO_3^-(aq) + H_2O_2(aq) \rightarrow HOSOO_2^-(aq) + H_2O(aq),$$

$$HOSOO_2^-(aq) + HA(aq) \rightarrow 2H^+(aq) + SO_4^=(aq) + A^-(aq),$$

in which HA is H_3O^+ or a suitable weak acid in the solution. According to this mechanism, the reaction occurs via a nucleophilic displacement by H_2O_2 on $HOSO_2^-$ to form a peroxymonosulfurous acid intermediate, $HOOSO_2^-(aq)$, which then undergoes a rate-determining rearrangement assisted by H_3O^+ or HA.

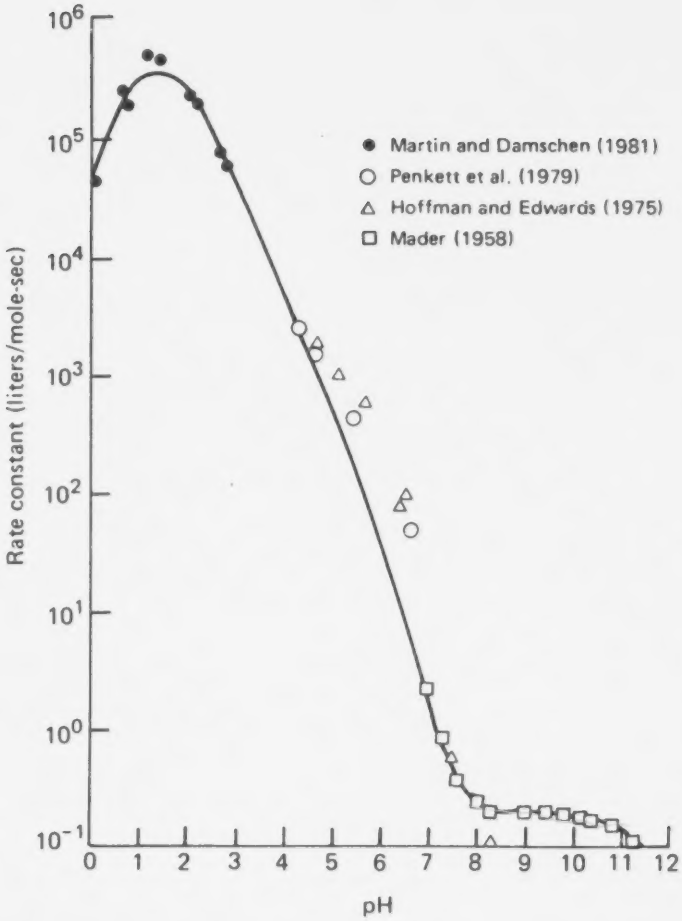


FIGURE A.8 Rate constant (k) for the bisulfite H_2O_2 reaction as a function of pH from the expression $-d[S(IV)]/dt = k[H_2O_2][S(IV)]$ with the effect of buffer removed and all results converted to 25°C. SOURCE: Martin (1983).

The oxidation of SO_2 by O_3 in solution has been studied by several groups. Available data are summarized in Figure A.9 (Martin 1983). For aqueous solutions with pH in the range 1-3, the rate of oxidation via ozone follows

$$-\frac{d[O_3(aq)]}{dt} = 1.9 \times 10^4 [H^+(aq)]^{-1/2} [O_3(aq)][S(IV)]$$

$$\text{mole liter}^{-1}\ \text{sec}^{-1}. \quad (69)$$

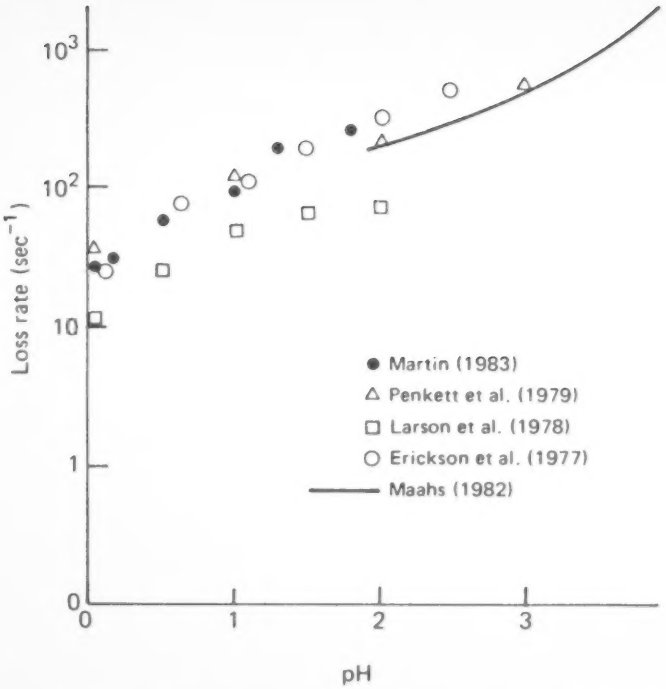


FIGURE A.9 Pseudo-first-order rate constant for ozone loss as a function of pH in the reaction of ozone with bisulfite with initial $[O_3] = 1 \times 10^{-4}$ and $[S(IV)] = 5 \times 10^{-4}$ M. SOURCE: Martin (1983).

For pH between 3 and 6.5, the rate law has this form:

$$-\frac{d[O_3(aq)]}{dt} = 4.19 \times 10^5 [O_3(aq)][S(IV)] + 100[H^+(aq)]^{-1}[S(IV)][O_3(aq)] \quad \text{mole liter}^{-1}\ \text{sec}^{-1}. \quad (70)$$

The inverse dependence of the rate of the O_3-S(IV) reaction on $[H^+(aq)]$ decreases the potential importance of the O_3 reaction with acidic solutions characteristic of the troposphere. The apparent activation energy of the reaction is 8.2 kcal/mole (Maahs 1982).

There is a strong link between the gas-phase and liquid-phase atmospheric chemistry related to acid precipitation. The major contributors to solution-phase oxidation of S(IV) in the troposphere, presumed to be H_2O_2 and O_3, are both products of the homogeneous

gas-phase reactions. Hence the gas-phase and liquid-phase pathways of acid generation in the troposphere are not independent.

The major sources of H_2O_2 in the gaseous troposphere are reactions (71) and (73), involving the HO_2 and the hydrated HO_2 radicals:

$$2HO_2 \rightarrow H_2O_2 + O_2, \tag{71}$$

$$HO_2 + H_2O \rightarrow H_2O{\cdot}HO_2, \tag{72}$$

$$H_2O{\cdot}HO_2 + HO_2 \rightarrow H_2O_2 + H_2O + O_2. \tag{73}$$

The complex $H_2O{\cdot}HO_2$ represents only a few percent of the total HO_2 concentration in the atmosphere; however, the inequality of the rate constants, $k_{73} > k_{71}$, ensures that its role in H_2O_2 formation is not insignificant. Of course, if water is available to gaseous H_2O_2, this compound will end up largely in the aqueous phase because of the large Henry's law constant for H_2O_2 in water (7.1×10^4 M atm^{-1} at 25°C). In the polluted atmosphere, there is strong competition for the HO_2 radicals through reactions (27), (74), (75), and possibly others.

$$HO_2 + NO \rightarrow HO + NO_2, \tag{27}$$

$$HO_2 + CH_2O \rightleftharpoons HO_2CH_2O \rightarrow O_2CH_2OH, \tag{74}$$

$$HO_2 + O_2CH_2OH \rightarrow HO_2CH_2OH + O_2. \tag{75}$$

The efficiency with which H_2O_2 is generated in the polluted troposphere through (71) and (73) is a complex function of [NO] and [CH_2O] as well as the concentrations of CO, hydrocarbons (RH), and aldehydes (RCHO) from which the HO_2 radical is derived. [See reactions (21), (22), (30), and others.] Simulations of the complex chemistry of the polluted troposphere show that with a highly polluted air mass ([NO_x] = 100 ppb, [RH] = 500 ppb, [RCHO] = 30 ppb), H_2O_2 development is unimportant until the [NO] has been depleted late in the day (Calvert and Stockwell 1983). However, for conditions more typical of a nonurban air mass, where it is not uncommon to have very low NO_x levels (≈1 ppb) and somewhat higher RH and RCHO concentrations in this circumstance, generation of H_2O_2 can begin early in the day. When [NO_x] is very high (100 ppb) and [RH] and [RCHO] are low initially ([RH] = 50 ppb, [RCHO] = 3 ppb),

little H_2O_2 forms. However, with small levels of all the pollutants, H_2O_2 formation in (71) and (73) can be significant. In theory, the amount of H_2O_2 formed in these gas-phase reactions can often be sufficient to oxidize a large fraction of the bisulfite usually present in cloud water and precipitation.

Production of O_3 in the troposphere follows a somewhat different pattern. Its development is most favored in the highly polluted atmosphere, although significant concentrations (0.08 ppm) may be generated in relatively clean urban and rural atmospheres. The Henry's law constant for O_3 in water (0.01 M atm^{-1} at 25°C) and normal concentrations of O_3 found in the relatively clear troposphere (30-60 ppb) ensure that the aqueous droplets in the atmosphere will contain ozone concentrations of about 3-6 x 10^{-10} M.

These limited observations suggest that conditions of low NO_x and high hydrocarbon and aldehyde impurity levels favor H_2O_2 formation, whereas those of relatively high NO_x, RH, and RCHO favor high O_3 generation rates. If homogeneous air masses containing preformed H_2O_2 and O_3 encounter cloud water, rainwater, high aqueous acid aerosol levels, etc., then solution phase pathways for H_2SO_4 formation are favored as well. Because the optimum impurity levels for generation of O_3 can be very different from those for H_2O_2 formation, the components of the H_2O_2 and O_3 oxidation of SO_2 (HSO_3^-) in the solution phase will not proceed exactly in phase with fixed fractions occurring by each pathway. The rates of SO_2 oxidation in the homogeneous gas-phase oxidation and those of the solution-phase reactions will depend on two quite different, complex functions of pollutant concentrations. However, it is clear that the two processes are not entirely independent.

Other interesting possibilities for the oxidation of S(IV) to sulfuric acid in cloud water have been suggested but remain untested. Chameides and Davis (1982) studied theoretically the contribution to aqueous-phase chemistry of HO and HO_2 gas-phase radicals scavenged by and incorporated into cloud droplets during daylight. A seemingly logical series of chemical reactions involving these species and their products was suggested, and calculations indicated that for small water droplets (<20 μm) this scavenging process may be a very important source of oxidant (H_2O_2, HO_2, HO) for acid development provided that the sticking coefficent for these species impinging upon the water droplets is greater than about 10^{-3}.

Both NO_2 and HONO can oxidize sulfite in dilute solutions. Martin et al. (1981) report that $HONO_2$ does not react significantly for any of the pH conditions employed. Lee and Schwartz (1981) and Schwartz and White (1982) estimated the oxidation rate of $NO_2(aq)$ with S(IV) to be 2.4×10^{-6} M/h at pH 5 and $P_{SO_2} = P_{NO_2} =$ 10 ppb in the gas phase. Their preliminary data suggest that for relatively high NO_2 and SO_2 pollution levels the acid generation from the NO_2-SO_2-H_2O(liq) system could be significant.

In laboratory experiments, nitrous acid can oxidize sulfite at a reasonably fast rate. Martin et al. (1981) report that the rate law for this system is given by

$$\frac{d[S(IV)]}{dt} = 142[H^+(aq)]^{1/2}[N(III)][S(IV)]$$

$$\text{mole liter}^{-1}\ \text{sec}^{-1}. \quad (76)$$

The rate constant data are summarized in Figure A.10. The nitrogen-containing product of the reaction up to pH 3.5 was N_2O. Above this pH, hydroxylamine disulfonate ($HON(SO_3)_2^{=}$) forms. Further reactions of this species occur to give hydroxylamine and nitrous acid. At the very low levels of gaseous HONO in the atmosphere both anticipated theoretically and observed experimentally, it is not likely that this reactant will contribute significantly to the oxidation of S(IV) in the troposphere.

The truly uncatalyzed oxidation of sulfite solutions by oxygen is very slow, and there is some question whether small impurities of metal ions such as iron may not account entirely for observed "uncatalyzed" rates (Martin 1983).

$$O_2(aq) + 2HSO_3^-(aq) \rightarrow 2H^+(aq) + 2SO_4^{=}(aq).$$

Both iron (Fe^{3+}) and manganese (Mn^{2+}) ions catalyze the reaction effectively. The rate law derived by Martin (1983) for the iron-containing system is given by

$$\frac{d[SO_4^{=}(aq)]}{dt} = 0.82[H^+(aq)]^{-1}[Fe^{3+}][S(IV)]$$

$$\text{mole liter}^{-1}\ \text{sec}^{-1}. \quad (77)$$

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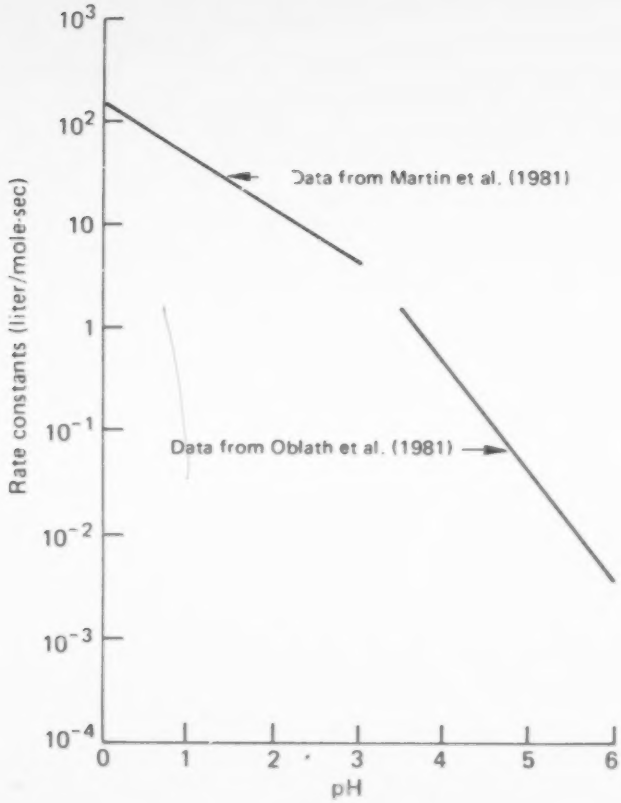

FIGURE A.10 Rate constant (k) for the reaction of HONO with bisulfite as a function of pH in the expression $d[S(IV)]/dt = k\,[N(III)]\,[S(IV)]$. SOURCE: Martin (1983).

These data and those of previous workers are summarized in Figure A.11. An increase in $[H^+(aq)]$ typical of a reacting mixture in cloud water or rainwater tends to lower the rate of this reaction pathway. In the high pH range (pH > 4), a condition uncommon for most natural atmospheric situations, ferric ion concentrations fall below 10^{-8} M, and the inconsistency of the rate laws observed in the laboratory for this system in this pH range may reflect the unrecognized problem of iron ion removal as a precipitate.

Catalysis of sulfite oxidation by manganese ions is governed by different mechanisms and rate laws depending on the concentrations of reactants. Martin (1983) reported the following expression for the [S(IV)] regime from 10^{-3} to 10^{-4} M:

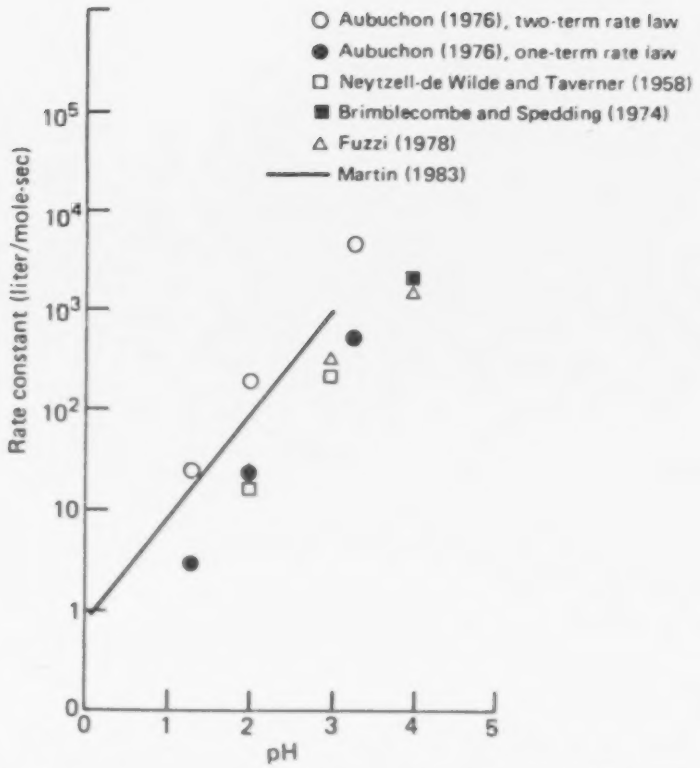

FIGURE A.11 Rate constant (k) for the iron-catalyzed oxidation of bisulfite as a function of pH in the expression $d[S(IV)]/dt = k\,[Fe^{3+}]\,[S(IV)]$ with data corrected to 25°C. SOURCE: Martin (1983).

$$\frac{d[SO_4^{=}(aq)]}{dt} = 4.7[H^+(aq)]^{-1}[Mn^{2+}(aq)]^2$$

$$\text{mole liter}^{-1}\ \text{sec}^{-1}. \quad (78)$$

The rate is independent of [S(IV)] for these conditions. This and other estimates of the rate constants for this reaction are shown as a function of pH in Figure A.12. The activation energy for the reaction is 27.3 kcal/mole (Hoather and Goodeve 1934a,b). At low [S(IV)] (less than 10^{-6} M), Martin (1983) finds the rate law to be

$$\frac{d[S(IV)]}{dt} = 25[H^+(aq)]^{-1}[Mn^{2+}][S(IV)]$$

$$\text{mole liter}^{-1}\ \text{sec}^{-1}. \quad (79)$$

For the entire range of concentration the rate is inversely proportional to $[H^+(aq)]$. For the conditions of relatively high concentrations of Fe^{3+} or Mn^{2+} in atmospheric precipitation or urban fogs (Hoffman and Jacob 1983), the rates of S(IV) oxidation through these catalyzed reactions can be very significant. These conditions usually do not prevail in the free troposphere in nonurban areas, however.

Barrie and Georgii (1976) found that when both Fe^{3+} and Mn^{2+} ions are present in sulfite solutions, the rate of sulfite oxidation is enhanced over that expected from the sum of the Mn^{2+}-S(IV) and Fe^{3+}-S(IV) rates alone. Martin (1983) studied these systems in detail and confirmed this finding. For the combined Fe-Mn system the rate of sulfite oxidation was typically 3 to 10 times faster than that anticipated from the sum of the independent rates in the individual systems.

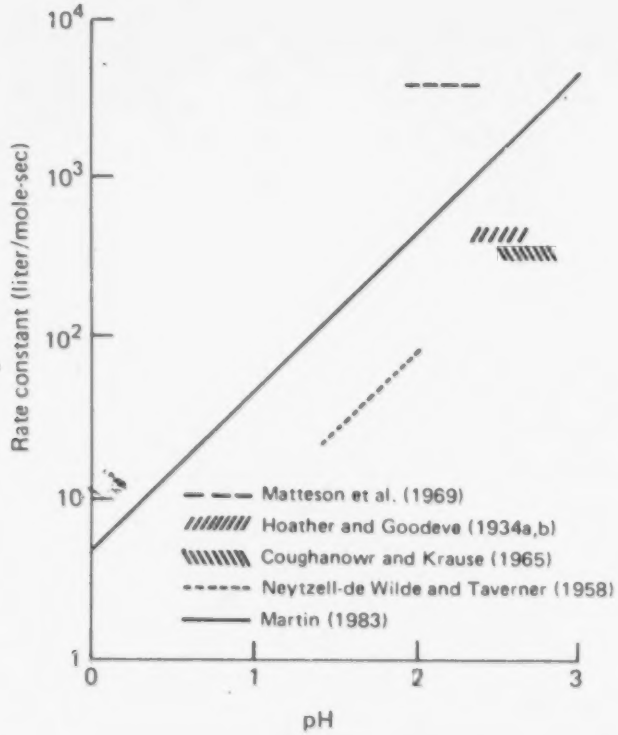

FIGURE A.12 Rate constant (k) for the Mn^{2+} ion catalyzed oxidation of bisulfite as a function of pH in the expression $d[S(VI)]/dt = k[Mn^{2+}]^2$. SOURCE: Martin (1983).

Ions of other metals such as Cu and Co are much less effective catalysts for sulfite oxidation than Mn and Fe for the low values of pH encountered in the environment (Martin 1983).

The mechanism by which the catalytic reactions occur has been studied by Hoffman et al. (1982) and others. A basic hypothesis being tested by Hoffman et al. is that the presence of certain trace metals in atmospheric aerosols and the occurrence of H_2O_2, HO, and HO_2 may be interrelated phenomena. Hoffman et al. illustrate this notion by the following mechanistic sequence:

$$Cu^+(aq) + O_2(aq) \rightleftharpoons CuO_2^+(aq),$$

$$CuO_2^+(aq) + H^+(aq) \rightarrow Cu^{2+}(aq) + HO_2(aq),$$

$$Cu^+(aq) + HO_2(aq) \rightarrow Cu^{2+}(aq) + HO_2^-(aq),$$

$$H^+(aq) + HO_2^-(aq) \rightleftharpoons H_2O_2(aq).$$

For example, in atmospheric systems, Cu^+, Co^+, and Fe^{2+} could be generated by photoinduced reduction of Cu^{2+}, Co^{2+}, and Fe^{3+}.

$$Cu^{2+}(aq) + h\nu \rightarrow Cu^{2+*}(aq),$$

$$Cu^{2+*} + H_2O(aq) \rightarrow Cu^+(aq) + HO(aq) + H^+(aq).$$

Subsequent catalytic decomposition of the intermediate hydrogen peroxide may produce HO and additional HO_2 free radicals. This hypothetical sequence of reactions could proceed according to the mechanism suggested for the classical "Fenton's reagent" reaction for the decomposition of H_2O_2. This mechanism involves a catalytic couple for Fe^{2+} and Fe^{3+} and a chain reaction that may proceed as follows:

$$Fe^{2+}(aq) + H_2O_2(aq) \rightarrow Fe(OH)^{2+}(aq) + HO(aq),$$

$$Fe^{3+}(aq) + H_2O_2(aq) \rightarrow Fe^{2+}(aq) + H^+(aq) + HO_2(aq),$$

$$Fe^{3+}(aq) + HO_2(aq) \rightarrow Fe^{2+}(aq) + H^+(aq) + O_2(aq),$$

$$HO(aq) + H_2O_2(aq) \rightarrow H_2O(aq) + HO_2(aq),$$

$$HO_2(aq) + H_2O_2(aq) \rightarrow O_2(aq) + H_2O(aq) + HO(aq).$$

The reactive intermediates, H_2O_2, HO, and HO_2, in the overall sequence described in the equations may oxidize S(IV) to H_2SO_4.

Hoffman et al. (1982) postulate another mechanism by which transition metals or transition metal complexes may catalyze the autooxidation of trace-level hydrocarbons present in cloud droplets. For a generalized hydrocarbon (RH) a possible sequence is as follows:

$$M^{2+}(aq) + O_2(aq) \rightarrow M^{3+}\text{-}O_2^-(aq),$$

$$M^{3+}\text{-}O_2^-(aq) + RH(aq) \rightarrow M^{2+}(aq) + R(aq) + HO_2(aq),$$

$$M^{3+}\text{-}O_2^-(aq) + RH(aq) \rightarrow M^{3+}\text{-}O_2H + R^-(aq),$$

$$M^{3+}\text{-}O_2^-(aq) + RH(aq) \rightarrow M^{3+}(aq) + R^-(aq) + HO_2(aq)$$
$$\rightarrow M^{3+}(aq) + R(aq) + HO_2^-(aq),$$

$$HO_2^-(aq) + H^+(aq) \rightarrow H_2O_2(aq).$$

There is some field evidence for such reactions in the atmosphere; rainwater analyses from Norway show significant numbers of alkanes, polycyclic aromatic hydrocarbons, fatty acids and esters, benzoic acids, etc.

The interesting facet of this reaction scheme is the production of peroxide in the cloud droplet by gas-phase scavenged hydrocarbon products. Hoffman et al. suggest that complexes of $Fe^{2+,3+}$, $Mn^{2+,3+}$, $Co^{2+,3+}$, and $V^{3+,4+}$ should be the most effective catalysts for the autooxidation of sulfite. They also showed that certain metal-catalyzed reactions of sulfite may be enhanced by a photoassisted pathway. The coupling of photolytic and metal-catalyzed processes is also consistent with the observed difference between daytime and nighttime SO_2 conversion rates. Dissolved organic molecules can act as competitive complexing agents for metals. The presence of complexing agents of this type will accelerate the dissolution of Fe_2O_3 and MnO_2, which are the likely sources of soluble iron and manganese in cloud droplets.

Another area of potential importance is that involving solid catalysts. Chang et al. (1978, 1981), and Brodzinski et al. (1980) showed that carbon can be an important catalyst for oxidation of sulfite in urban smog. They found from experiments at 20°C that the following rate expression decribed their results:

$$\frac{d[S(IV)]}{dt} = 1.69 \times 10^{-5}[C_x]\,[O_2(aq)]^{0.69} \cdot \left\{ \frac{1.5 \times 10^{12}[S(IV)]^2}{1 + 3.06 \times 10^{6}[S(IV)] + 1.5 \times 10^{12}[S(IV)]^2} \right\} \quad (80)$$

The rates are in moles per liter per second, concentrations of soluble species are in moles per liter, and $[C_x]$ is the suspended carbon content of the suspension in grams per liter. The rate is independent of the pH (1.45-7.5), but it is dependent on the type of carbon (graphite, soot, etc.) employed and the surface area of the carbon particles.

Comparison of Reaction Pathways for Solution-Phase Oxidation of SO_2

The potential contributions of the various reactions we have considered here to solution-phase chemistry are compared for several realistic atmospheric conditions in Figure A.13. To obtain Figure A.13, Martin (1983) assumed that there are no limitations due to mass transport rates. The rate constants presented here have been applied to a hypothetical cloud containing 1 ml of liquid water per cubic meter of air at 25°C. The concentrations of reactants assumed in the calculation are listed on the figure. Rates for other impurity concentrations may be derived easily by recognizing that most of the processes are first order in each species so that the conversion rates are independent of SO_2 and linearly proportional to the concentration of the other species. The following are exceptions: (1) The mechanism for manganese-catalyzed oxidation changes at pH 4, because the equilibrium concentration of S(IV) goes above 10^{-6} M at higher values of pH; the shape of the curve is sensitive to the assumed concentrations of sulfur and manganese. (2) Oxidation by carbon catalysis is nonlinear in sulfur, and the curve of Figure A.13 will change in shape if the sulfur concentration is changed. (3) The upper portion of the curve for iron catalysis is nonlinear in sulfur.

For iron-catalyzed oxidation at high pH, Martin used the result of Brimblecombe and Spedding (1974) converted to a third-order rate. The dotted part of the curve between pH 3 and 5 is very uncertain, and there may be local maxima in this region. No synergism has been included in deriving the data of Figure A.13.

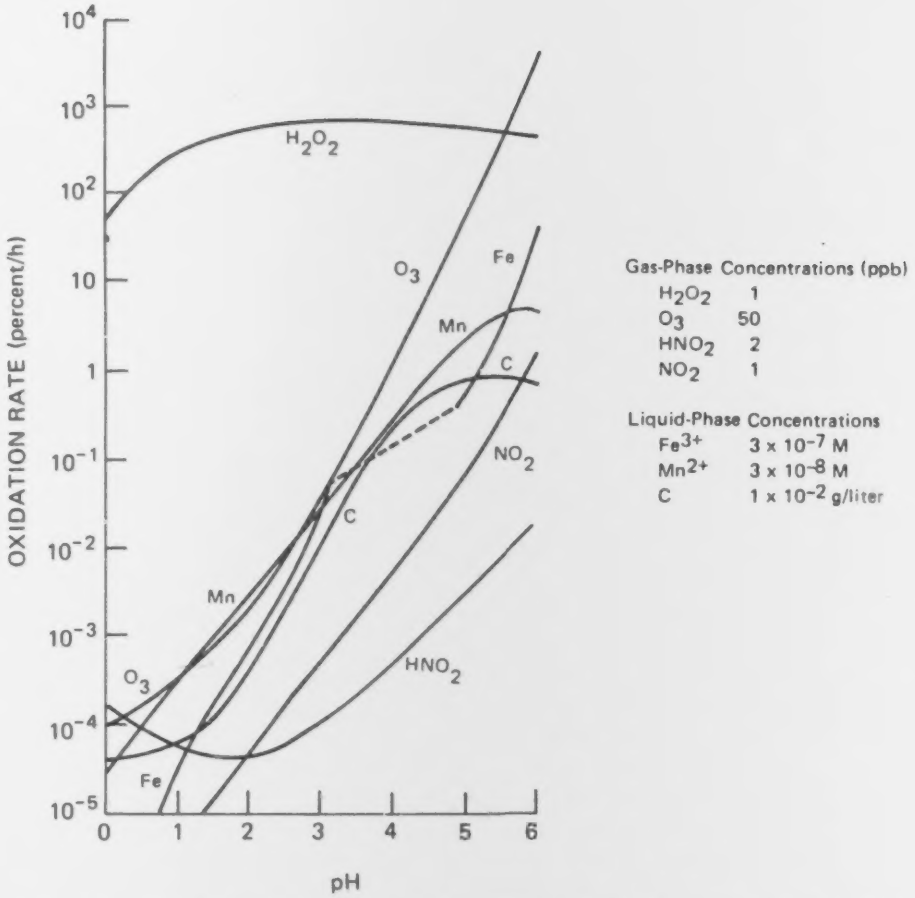

FIGURE A.13 Theoretical rates of liquid-phase oxidation of SO_2 assuming 5 ppb of SO_2, 1 ml/m^3 of water in air, and concentrations of impurities as shown. SOURCE: Martin (1983).

For the hydrated NO_2 oxidation, Martin used the Henry's law constant and rate constants of Schwartz and White (1982) and Lee and Schwartz (1981). The concentrations of NO_2, HONO, and NO are related thermodynamically, and so they may not be regarded as completely independent.

The trend of rising conversion rates with increased pH results from either the rising equilibrium concentration of sulfur (IV) or the sensitivity of the rate constants to pH, or both. The relative conversion rate increases with rising concentrations of sulfur (IV) in this range of pH values and for this amount of liquid. H_2O_2, which

undergoes an acid-catalyzed reaction, is the only oxidant for which the rate dependence on $[H^+]$ compensates for the decreased solubility of SO_2 with increased $[H^+]$; hence the rate of oxidation by H_2O_2 is nearly independent of pH.

Martin emphasized that the relative positions of the curves in Figure A.13 differ for differing assumptions about the composition of the droplets. For example, an aerosol with less liquid water will, as a rule, have higher concentrations of nonvolatile species such as carbon, iron, and manganese but a smaller reaction volume, so the relative positions of the curves may differ in this situation. The relative positions will also be different at night, when the concentration of photochemically derived oxidants drops. At temperatures below 25°C, the rate constants are lower in accordance with the activation energies of the given reactions, and the Henry's law coefficients are higher. The two effects act in opposite directions. In some cases, such as that for H_2O_2 oxidation, the net rate rises as the temperature falls (at constant gas-phase concentration). In other cases, such as for iron catalysis, the net rate falls with temperature.

The concentrations of reactants used in deriving Figure A.13 are representative of those that might be anticipated in the atmosphere. Oxidation by H_2O_2 dominates all reactions for conditions of low pH. Oxidation rates can be greater than 100 percent/h. The rate for oxidation by ozone varies from about 10 percent/h at pH 4.5 to about 1 percent/h at pH 4.0. The contributions from the Fe^{3+}, Mn^{2+}, and carbon-catalyzed reactions (for the conditions specified) are below 1 percent/h for solutions of pH < 4.5. Oxidation by NO_2 and HONO is even less significant under these conditions.

An additional influence on the rates and kinetics of sulfite oxidation in clouds and rainwater can arise from the complexation of HSO_3^-(aq) by aldehydes scavenged by the droplets. The common gaseous products of atmospheric oxidation of the impurity hydrocarbons, CH_2O, CH_3CHO, CH_3COCH_3, and possibly other carbonyl compounds, can in principle complex with HSO_3^-. The result of such interactions could increase the solubility of SO_2 in the droplet and conceivably retard sulfite oxidation by the oxidants.

Although there is significant theoretical evidence that H_2O_2 may be the most important oxidizing agent for acid generation in cloud water and rain, unambiguous

experimental measurements of H_2O_2 levels in air and in cloud water and rain have not been possible to date. Peroxide development in the sampling train and lack of selectivity of the luminol technique employed in previous work prevents a firm conclusion about the H_2O_2 levels in the atmosphere and in precipitation (Heikes et al. 1982, Zika and Saltzman 1982). Theoretical simulations of the atmospheric chemistry of a mixture of reactants in a highly diluted urban atmosphere show that H_2O_2 generation through reactions (71) and (73) can provide substantial levels of H_2O_2, CH_3O_2H, and other hydroperoxides in the gas phase. If these species are absorbed into aqueous aerosols, cloud water, or rainwater, for example, as is probable for H_2O_2 in view of its very high Henry's law constant, then oxidation by H_2O_2, and possibly other peroxides, can be most significant. The possible roles for peroxyacetylnitrate, peroxynitric acid, CH_3O_2H, and other peroxides in solution-phase sulfite oxidation remain to be evaluated.

Solution-Phase Generation of Nitric and Nitrous Acids in the Troposphere

There is some evidence of the formation of $HONO_2$ in clouds and rainwater. Recently, both theory and experiment suggest that $HONO_2$ may be formed rapidly from a combined gas-phase/liquid-phase process. Through simulation, Heikes and Thompson (1981) have suggested that N_2O_5 generated by O_3-NO_2 reactions (15) and (16) may be scavenged effectively by H_2O droplets to form $HONO_2$ in clouds or rainwater.

$$NO_2 + O_3 \rightarrow O_2 + NO_3, \tag{15}$$

$$NO_3 + NO_2(+M) \rightleftharpoons N_2O_5(+M), \tag{16}$$

$$N_2O_5 + H_2O(liq) \rightarrow 2H^+(aq) + 2NO_3^-(aq) \tag{81}$$

A preliminary report of the experimental observation of this process has been made recently by Gertler et al. (1982). An evaluation of the contribution of this mechanism to the total [$HONO_2$] found in the atmosphere is not now possible.

Lee and Schwartz (1981) studied the rates of reactions (82) and (83):

$$2NO_2(g) + H_2O(liq) \rightarrow 2H^+(aq) + NO_3^-(aq) + NO_2^-(aq), \quad (82)$$

$$NO(g) + NO_2(g) + H_2O(liq) \rightarrow 2H^+(aq) + 2NO_2^-(aq). \quad (83)$$

They found that at the lower partial pressures characteristic of moderately polluted atmospheres the rates are slow (10^{-9} to 10^{-8} M/h). Unless high partial pressures (e.g., 10^{-7} atm) of these gases are maintained in contact with liquid water for substantial periods of time (tens of hours), reactions (82) and (83) cannot represent a substantial source of atmospheric acidity. Lee and Schwartz found no evidence of Fe^{2+} ion catalysis of these reactions, but they suggested that possibly other metal ions could enhance these rates. There is no evidence related to this possibility now available.

Although significant uncertainty remains concerning the source of HNO_3 in clouds and rainwater, the limited evidence currently available favors the probable importance of the formation of N_2O_5 in (15) and (16) followed by its reaction in water droplets to form HNO_3.

SUMMARY

In summary, a wide variety of interrelated homogeneous gas-phase, solution-phase, and heterogeneous chemistry may result ultimately in oxidation of SO_2 to sulfuric acid and NO_x to nitric acid. The homogeneous gas-phase oxidation of SO_2 by the HO radical and the solution-phase oxidation of S(IV) through H_2O_2, O_3, and possibly other species appear to be the major sources of H_2SO_4. In the cloud-free, ambient, sunlight-irradiated troposphere, nitric acid is probably generated largely by the reaction of HO radicals with NO_2. Both $HONO_2$ and H_2SO_4 produced in the gas phase can be scavenged effectively by cloud water and precipitation. NO_2 may be oxidized to $HONO_2$ if sufficient O_3 and NO_2 are present. Following its gas phase generation, N_2O_5 may be scavenged effectively by water droplets to form $HONO_2$.

All the various pathways that lead to the oxidation of SO_2 and NO_x are coupled by common products and reactants that can directly and indirectly influence the rates of reaction by the other pathways. In general the homogeneous gas-phase reactions can lead to maximum daylight rates of acid formation of a few percent per hour for SO_2 and 20-30 percent/h for NO_2. Solution-phase reactions involving H_2O_2 and O_3 can in principle

convert SO_2 to H_2SO_4 in cloud water and precipitation at much higher rates (as high as 100 percent/h) for concentrations of H_2O_2 and O_3 in the troposphere that reasonably could result from the normal homogeneous reactions characteristic of atmosphere chemistry during daylight.

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Appendix B Transport and Dispersion Processes

Over the past 50 years scientists have been concerned with the transport of materials by the atmosphere. Substances of interest have included volcanic debris, Saharan dust, radioactive fallout, and industrial pollutants. The type of gas or particulate matter, their physical and chemical properties, the vigor of the atmospheric flow, and other factors help to determine how and where the material is finally deposited on the Earth's surface. This appendix treats only the physical transport of materials in the atmosphere. The effects of chemical transformation and scavenging by clouds and aerosols are discussed in detail in other appendixes.

The term transport encompasses the processes by which a substance or quantity is carried past a fixed point or across a fixed plane. In the atmosphere, the substances or quantities of interest include air parcels, gaseous impurities, suspended particles, and moisture (Huschke 1959).

CLASSIFICATION OF TRANSPORT PHENOMENA

Atmospheric motion and transport phenomena are extremely complex in both horizontal and vertical dimensions, with thermal layering, shear turbulence, convection, variation of boundary characteristics, and so on. Because of these complexities, meteorologists have devised an ordering of the various atmospheric phenomena. One way to approach the ordering is on the basis of spatial scale. After release, a given material diffuses during transport, coming under the influence of larger-scale motions as it moves farther from the source. To classify transport behavior, four scales have been defined: local, meso,

synoptic, and global. The local scale is defined as being on the order of the vertical dimension of the planetary boundary layer within which pollutants are typically emitted. This dimension is the order of a kilometer, and the time scale on which phenomena take place on this dimension is on the order of tens of minutes. The next largest spatial scale is the mesoscale, which extends up to several hundred kilometers and has an associated time scale of the order of a day (about the time needed for a mean horizontal transport of several hundred kilometers). Mesoscale effects include the diurnal variability of the planetary boundary layer and, therefore, the dynamics of plumes. In the mesoscale, an individual plume from a power plant or urban complex of sources loses its identity by mixing with other plumes or by diluting into the background. The synoptic scale is on the order of 1000 km, with transport times of about 1 to 5 days. The hemispheric or global scale reflects intercontinental transport with times on the order of a week. The term "long-range transport" commonly refers to transport on the synoptic and global scales.

The prevailing winds in the lower troposphere transport and disperse atmospheric pollutants. A combination of the rotation of the Earth (Coriolis effect) and the existence of synoptic-scale pressure gradients in the atmosphere maintain the planetary or geostrophic winds. A number of perturbances near the Earth's surface, such as surface roughness, heat, and moisture fluxes, influence the local winds. The perturbed layer, called the planetary boundary layer (PBL), is of variable height ranging typically up to 3 km. Because most atmospheric pollutants are released in this layer, study of the PBL is vital to understanding local or mesocale transport as opposed to synoptic or global transport.

LOCAL AND MESOSCALE TRANSPORT

Mesoscale transport is usually confined to the planetary boundary layer or the lowest 3 km. Embedded in this region and closest to the ground is a highly dynamic layer termed the mixing layer. Here the local effects of mechanical and thermal turbulence can predominate. It is called the mixing layer because within it atmospheric turbulence very effectively and quickly mixes and dilutes any concentrated release of mass, momentum, or heat. In other parts of the atmosphere, dilution may be slow. The

mixing layer typically undergoes a diurnal cycle rising to heights of 1 to 2 km in the day and lowering to 100 to 300 m at night. Thermal convection dominates in the day, and small-scale mechanical turbulence at night. Because of the efficient mixing during the day, pollutants are quickly moved to all areas of the mixing layer including the ground. On the other hand, elevated releases at night may be above the shallow mixing layer and can be transported independently.

In general, mesoscale mean winds dominate plume transport, but, depending on the strength of local turbulent eddies, the plume may also be spread horizontally and vertically. Another factor affecting pollutant transport is wind shear. Since thermal convection and mechanical drag of the ground diminishes with height, the geostrophic balance of forces varies with height and is maintained by increasing wind speed and veering in wind direction. Thus, vertically adjacent layers of air move at different speeds in different directions (shear). Wind shear may cause dispersion and dilution of atmospheric pollutants and becomes increasingly important as the range of transport increases.

In addition to this general picture of local and mesoscale transport, there are significant diurnal and seasonal variations in the boundary layer that affect transport on these two scales. Figure B.1 shows the different patterns for winter and summer. The major feature to notice is that for both periods there is a very stable nocturnal layer that extends to 300 m. However, during the daytime, mixing heights are much greater in summer than in winter when an elevated daytime inversion hinders vertical mixing.

Another factor to be considered on the mesoscale is the vertical profile of the horizontal wind speed. Diurnal and seasonal variations in the profile are affected by the vigor of the synoptic-scale flow. Winter is a period of frontal passages, whereas in summer weak anticyclonic systems tend to prevail (Figure B.2).

From the above discussion, it can be seen that the mesoscale transport and dilution of a given pollutant depend on whether its source is elevated or on the ground. For example, while most of the SO_2 is emitted from elevated point sources, NO_x emissions are more evenly distributed between elevated and ground-level sources. Thus on the average, elevated releases spend more of their mesoscale transport time decoupled from the ground, while near-ground releases maintain continuous

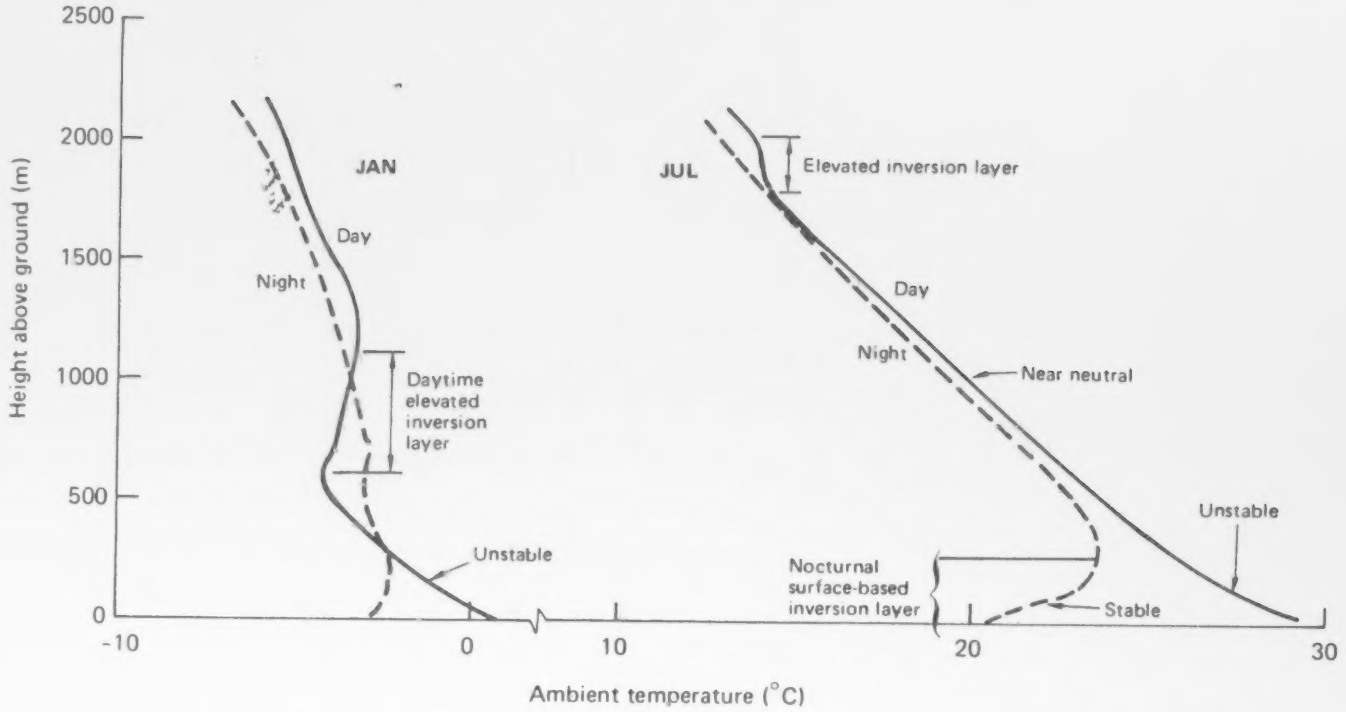

FIGURE B.1 Monthly average diurnal and seasonal variations of the vertical thermal structure of the planetary boundary layer at a rural site near St. Louis, Missouri, based on 1976 data. SOURCE: N. Gillani, Washington Unversity, St. Louis, Missouri, personal communication (1982).

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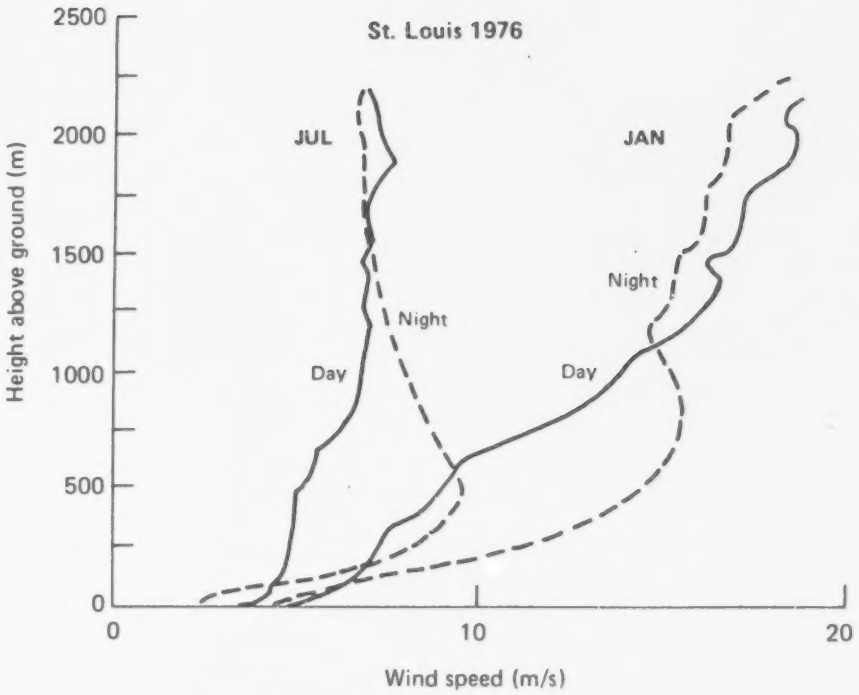

FIGURE B.2 Monthly average diurnal and seasonal variations of the vertical profiles of wind speed near St. Louis, Missouri, based on 1976 data. SOURCE: N. Gillani, Washington University, St. Louis, Missouri, personal communication (1982).

ground contact. This fact has a direct relation to the importance of diurnal and seasonal dry deposition and to some degree on the wet-deposition patterns.

While the main emphasis in acid deposition has been on the long-range transport of pollutants to remote areas, consideration of mesoscale transport and dispersion of pollutants of varying source types under varying flow conditions have an important bearing on how much of the emissions become available for long range transport and in what form. Important mesoscale factors such as release height and diurnal and seasonal variabilities must not be neglected in long-range transport modeling.

SYNOPTIC- OR CONTINENTAL-SCALE TRANSPORT

Synoptic transport of pollutants, especially acids and acid precursors, has been one of the major thrusts in acid deposition research. Numerous models have been

devised to investigate the long-range transport (up to 1000 km) and are reviewed in Chapter 5 of this report. Each of the models simulates the transport, transformation, and deposition of a given substance (sulfur compounds in this case). The main transport module in the models uses the synoptic winds, which are measured in the vertical dimension every 12 h by balloon soundings. The network of balloon soundings produces, unfortunately, sparser data coverage than the precipitation chemistry measurements over eastern North America. Considering the spacing of upper-air measurements, it is optimistic to expect the knowledge of the direction of the prevailing wind at an arbitrary location in space and time to be known to better than 5° about the "actual" advecting wind. When one calculates forward or back trajectories from these winds, there is an uncertainty in the cross-wind direction of 15 to 20 percent of the trajectory length for every timestop forward or backward in time. One would hope that such uncertainties and errors would cancel out when trajectories are calculated over many days and a climatology is established.

There are several key factors that determine the transmission of pollutants on the synoptic scale specifically over the North American continent. Already mentioned is the wind field. Clear patterns can be seen from a summary of the 1975-1977 data (R. Husar, Washington University, St. Louis, MO, personal communication, 1982). Conclusions are that (1) the general flow is from west to east with an important component northward from the Gulf of Mexico, (2) winter and fall have the highest speeds, (3) the southeastern United States is within a region of low mean velocity during late spring and summer, and (4) the Midwest exhibits very strong shear during summer and spring (Figure B.3).

It is important to note that winds above 1 to 2 km are not always important in the transport of surface releases, depending on the mixing depth. Also, well-mixed aged pollutants in the nocturnal stable layers may not always be re-entrained into the mixing layer the next morning. Contours of mixed depths (Figure B.4) provide some insight into the gross interaction of advecting winds and the depth of the mixing layer. However, synoptic temporal and spatial scales of interaction may be at least as important as the seasonal averages in determining the net transport of emissions. It is important to note that some of the well-mixed aged pollutants will ride over the

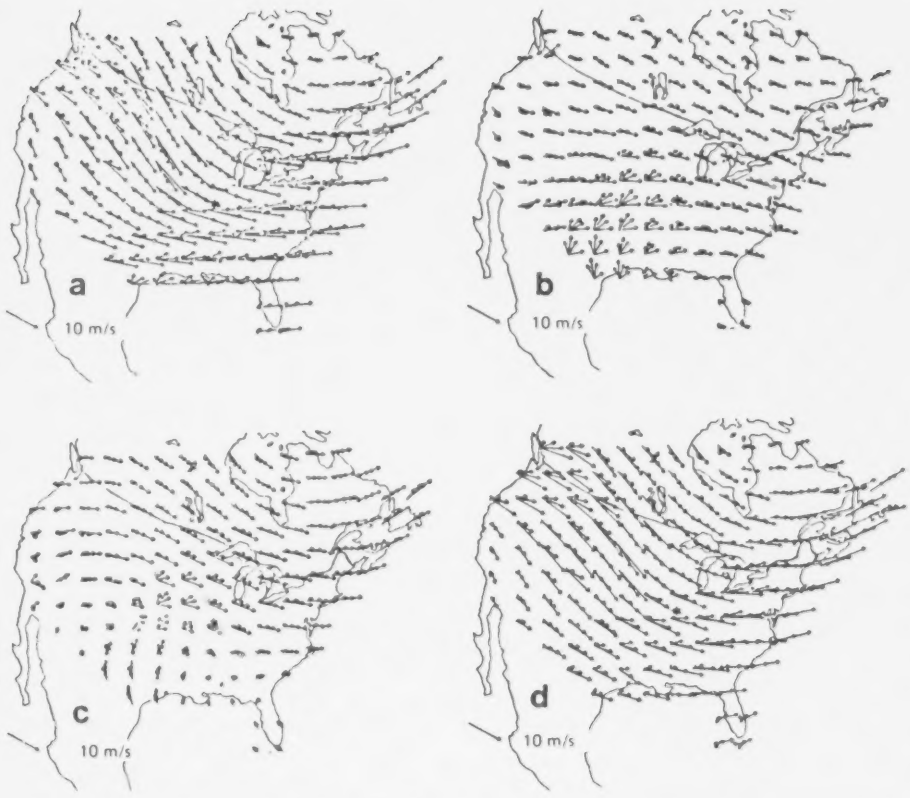

FIGURE B.3 Averages for 1975-1977 of winds in the layers 0-500, 500-1000, 1000-2000, and 2000-3000 m agl for the 000 and 1200 GMT soundings. Lower-level winds generally lie to the left and are of lower speed. a, January through March; b, April through June; c, July through September; and d, October through December. SOURCE: R. Husar, Washington University, St. Louis, Missouri, personal communication (1982).

daytime mixed layer when moving either from south to north or from west to east owing to lowered mixed depths along the trajectory. Parameterization of the vertical structure in the models is important for simulation of continental-scale transport over several days and thousands of kilometers.

Other vertical motions must be taken into account in long-range transport, although these are difficult to simulate properly. Vertical motions are important, for example, in transmission of pollutants across major physical barriers (for example, the Rocky Mountains), along warm and cold fronts, and near simple convective cells or clusters of cells. Also the vigor of motion of

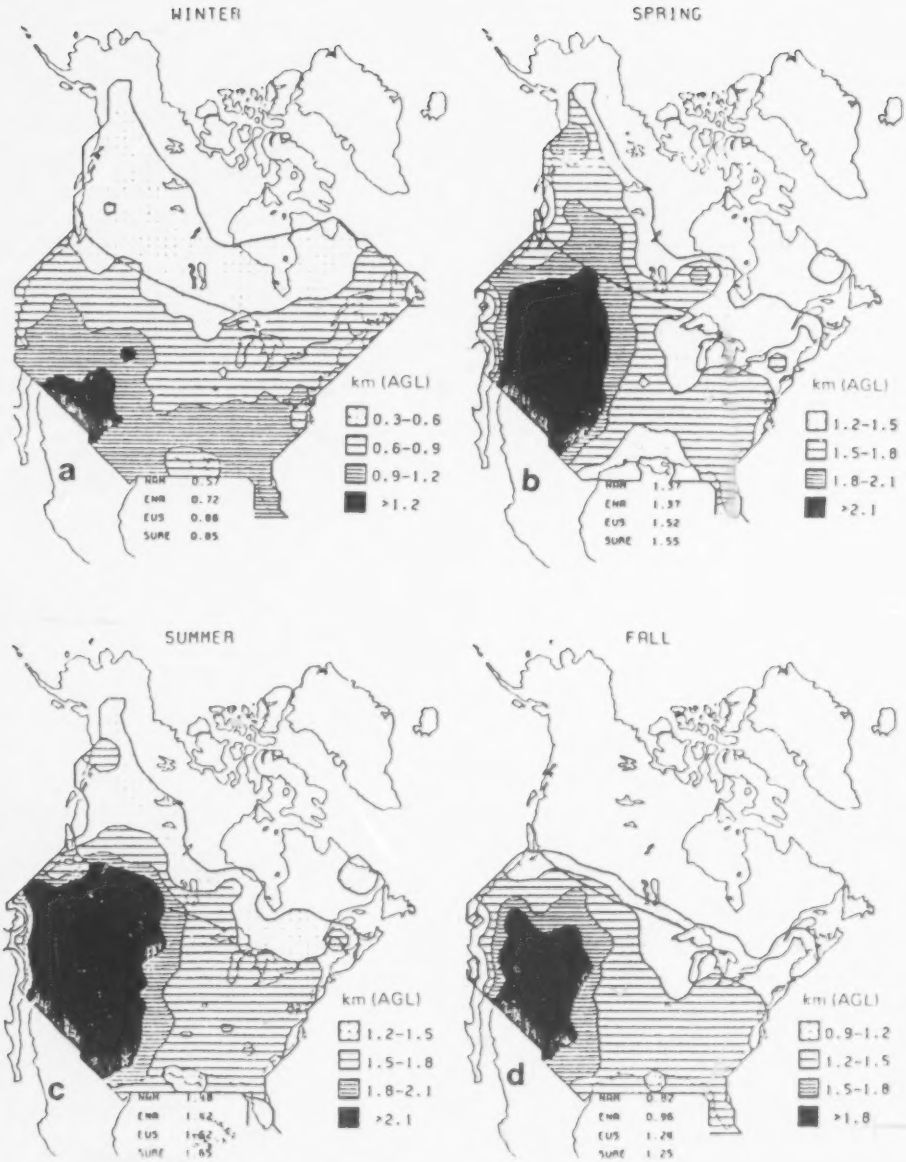

FIGURE B.4 Contour plots of maximum afternoon mixing depths by season, indicating qualitative patterns only. a, January through March; b, April through June; c, July through September; and d, October through December. SOURCE: Holzworth (1972) and Portelli (1977).

both cyclonic and anticyclonic systems can have an impact on accumulation of emissions. Korshover (1976) has pointed out that the south central United States is particularly subject to stagnating anticyclones, leading to lower ventilation of local and advected emissions.

Another factor critical to long-range transport is precipitation, which removes pollutants in a sporadic way. Trajectories from a source to a receptor will not establish the total mass transported if the air mass is likely to experience precipitation along the way. This removal depends on the type, intensity, and frequency of the precipitation. At present, the precipitation removal process is difficult to quantify over long transport paths.

Recently another important factor has been pointed out by Draxler and Taylor (1982). The authors showed that the spreading of emissions is dominated by the action of vertical wind shear acting in combination with the diurnal cycle of daytime mixing and nighttime layering of the atmosphere. Further work on the importance of this factor is being pursued.

In understanding the synoptic-scale transport, one should not lose sight of the fact that both local and mesoscale influences are important in continental transport. Thus to model the regional transport, the mesoscale must be adequately parameterized even if not explicity nested with that scale's simulation.

HEMISPHERIC OR GLOBAL TRANSPORT

Besides the fact that hemispheric transport involves greater distances and times than regional transport, there are important differences between the two scales. One fact is that the bulk of the global transport takes place over water. Because of the small changes in oceanic surface temperature, the planetary boundary layer over the oceans is relatively constant. Besides this, the oceans can be considered a homogeneous surface over large areas. Thus there are broad stretches of strong atmospheric inversions over cold water and other well-mixed regions over relatively warm water. One can expect that pollution within the boundary layer is subject to dry removal and that pollution that has been transported above the boundary layer will remain there until removed by precipitation processes or by large-scale subsidence.

The study of the movement of acidic and preacidic material from sources in North America to other receptor regions in the northern hemisphere has been undertaken in several cases. However, because of the lack of chemical and meteorological data over large stretches of the ocean, only crude estimates of this transport can be made. For example, the high acidity found in precipitation on the island of Hawaii could be partially explained by long-range transport from the west, where Japan would be the major source (Miller and Yoshigana 1981, Dittenhoefer 1982). In this study, a single trajectory model was useful in evaluating the transport patterns.

Another area of interest is the contribution of North American sources to Arctic haze. This issue has been raised more in reference to visibility or the modification of the radiation balance, since the Canadian and U.S. Arctic areas are deserts (100 mm per annum) with little wet deposition. The major transport path from eastern North America is a track around Greenland. Concentrations of pollutant aerosols in the Arctic show a definite wintertime peak when removal mechanisms are most inactive. Rahn and McCaffrey (1980) indicate that residence times of aerosol particles in the Arctic range from 2 to 3 weeks in the winter.

The transport of materials across the Atlantic has also been a topic of interest though not firmly established. Early estimates were made that North American contribution to sulfate in rain in Norway could be important. More recent studies in Bermuda indicate that trans-Atlantic transport of acid precursors is important to the acidity of precipitation on the island (Jickells et al. 1982). Further studies of this transport are being continued under a joint U.S.-Canada-Bermuda effort.

Recent studies of precipitation in remote areas of both the northern and southern hemisphere have shown the acidity of rain to be on the average lower than pH 5.0 (Galloway et al. 1982). The degree to which natural sources or long-range transport of man-made pollutants contribute to this remote acidity in precipitation remains to be seen. However, trajectory calculations to estimate the transport on a global scale will be a useful tool in such research.

CONCLUSIONS

Though the transport of materials in the atmosphere has been studied for a number of years, there is still much

that can be learned in applying this knowledge to the acid deposition problem. By and large, the vigor of the atmosphere in both the horizontal and vertical rules where the final deposition of a given pollutant will be.

ACKNOWLEDGMENTS

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Appendix C Atmospheric Deposition Processes

1. INTRODUCTION

In this appendix we present an overview of current scientific understanding about deposition phenomena, with the objectives of identifying key literature sources on this subject and providing the reader with the technical basis necessary for effective evaluation of the available literature. There are several important features of this subject, which should be noted at the outset. First, the ultimate deposition processes of interest are the end products of a complex sequence of atmospheric phenomena (cf. Figure 2.1). Deposition processes tend to reflect these preceding events strongly. Much of the material presented in this appendix therefore necessarily deals with the predeposition processes, which may act as important rate-influencing steps in the overall source-deposition sequence.

A second important feature of initial interest is the relative difference in states of our current understanding of wet- and dry-deposition phenomena. Wet deposition is comparatively simple to measure. As a consequence there exists a substantial and growing base of data on wet deposition from a variety of networks and field studies. Precipitation processes tend to be rather complicated, however, and currently a high level of uncertainty exists regarding their mathematical characterization.

Dry deposition, on the other hand, tends to be extremely difficult to measure, and the corresponding data set is relatively meager. Partly because of this fact most mathematical characterizations of dry-deposition processes have been quite simple in form. The tendency toward simplicity in most mathematical characterizations of dry deposition should not be taken to

imply that the physical processes themelves are simple. As a consequence of these differences the following sections on dry and wet deposition have somewhat different formats, with emphasis in each placed on areas of current major activity.

Finally, it should be noted that very little of the material presented in this appendix is new. A number of reviews of both wet and dry deposition have been presented during recent years, and the current treatment is merely an attempt to consolidate these efforts.* In view of this tendency toward redundancy, it is strongly recommended that the reader proceed directly to the indicated journal literature if more detailed pursuit of this subject is desired.

2. DRY-DEPOSITION PROCESSES

2.1 MECHANISMS OF DRY DEPOSITION

2.1.1 Introduction

The rate of transfer of pollutants between the air and exposed surfaces is controlled by a wide range of chemical, physical, and biological factors, which vary in their relative importance according to the nature and state of the surface, the characteristics of the pollutant, and the state of the atmosphere. The complexity of the individual processes involved and the variety of possible interactions among them combine to prohibit easy generalization; nevertheless, a "deposition velocity," v_d, analogous to a gravitational falling speed, is of considerable use. In practice, knowledge of v_d enables fluxes, F, to be estimated from airborne concentrations, C, as the simple product, $v_d \cdot C$.

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Particles larger than about 20-μm diameter will be deposited at a rate that is controlled by Stokes law, although with some enhancement due to inertial impaction of particles transported to near the surface in turbulent eddies. The settling of submicrometer-sized particles in air is sufficiently slow that turbulent transfer tends to dominate, but the net flux is often limited by the presence of a quasi-laminar layer adjacent to the surface, which presents a considerable barrier to all mass fluxes and especially to gases with very low molecular diffusivity. The concept of a gravitational settling velocity is inappropriate in the case of gases, but transfer is still often limited by diffusive properties very near the receptor surface.

Sehmel (1980b) presents a tabulation of factors known to influence the rate of pollutant deposition upon exposed surfaces. Figure C.2-1 has been constructed on the basis of Sehmel's list and has been organized to emphasize the greatly dissimilar processes affecting the fluxes of gases and large particles. Small, submicrometer-diameter particles are affected by all the factors indicated in the diagram; thus, simplification is especially difficult for deposition of such particles. In reality, Figure C.2-1 already represents a considerable simplification, since many potentially important factors are omitted. In particular, the emphasis of the diagram is on properties of the medium containing the pollutants in question; a similarly complicated diagram could be constructed to illustrate the effects of pollutant characteristics. For particles, critical factors include size, shape, mass, and wettability; for gases, concern is with molecular weight and polarization, solubility, and chemical reactivity. In this context, the acidity of a pollutant that is being transferred to some receptor surface by dry processes is a quality of special importance that may have strong impact on the efficiency of the deposition process itself.

Figure C.2-2 summarizes particle size distributions on a number, surface area, and volume basis. In this way, the three major modes are brought clearly to attention. The number distribution emphasizes the transient (or Aitken) nuclei range, 0.005-0.05-μm diameter, for which diffusion plays a role in controlling deposition. The area distribution draws attention to the so-called accumulation size range formed largely from gaseous precursors (0.05-2-μm diameter, affected by both diffusion and gravity). The remaining mode (2-50-μm

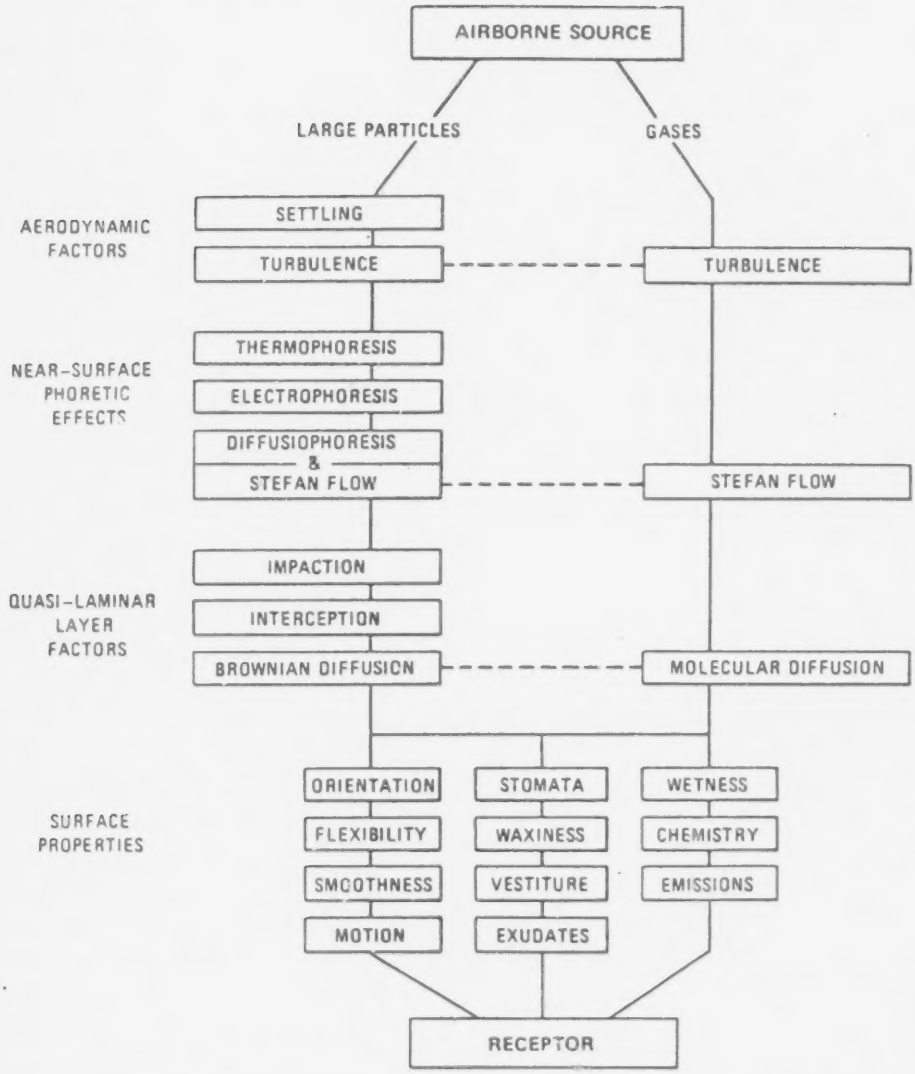

FIGURE C.2-1 A schematic representation of processes likely to influence the rate of dry deposition of airborne gases and particles. Note that some factors affect both gaseous and particulate transfer, whereas others do not. However, submicrometer particles are affected by all the factors that influence gases and large particles, and hence it is these "accumulation-size-range" aerosols that present the greatest challenge for deposition research.

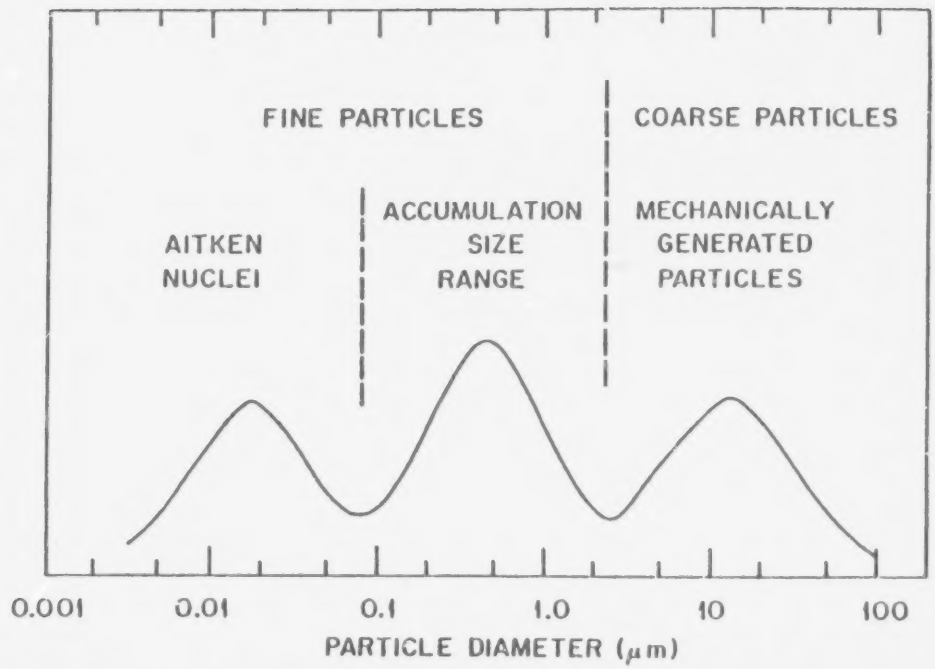

FIGURE C.2-2 A hypothetical particle-size spectrum, such as might be found downwind of an industrial complex. The smaller aerosols have gaseous precursors and are formed by condensation of exhaust gases and by atmospheric chemical reactions (typically oxidation), followed by growth due to particle coagulation. The larger particles are partly soil-derived, suspended by natural erosion and agricultural practices, and partly the direct result of the combustion of fossil fuels. Acidic aerosols are primarily in the smaller mode of the particle-size spectrum, whereas the larger mode contains material that might tend to neutralize the acidic deposition of the smaller particles. In evaluating the net input of acidity to a surface, it is critical that both size fractions and gaseous contributions be included.

diameter, most evident in the volume distribution) is the mechanically generated particle range for which gravity causes most of the deposition. In most literature, 2-μm diameter is used as a convenient boundary between "fine" and "coarse" particles.

Atmospheric sulfates, nitrates, and ammonium compounds are primarily associated with the accumulation size range. Figure C.2-2 demonstrates that very little acidic or acidifying material is likely to be associated with the coarse particle fraction in background conditions. However, the larger particles include soil-derived minerals, some of which can react chemically with airborne and deposited acids. Moreover, it has been

suggested that some of these larger particles may provide sites for the catalytic oxidation of sulfur dioxide (for example when the particles are carbon; Chang et al. 1981, Cofer et al. 1981). Little is known about the detailed chemical composition of large particle agglomerates. However it is accepted that their residence time is quite short (i.e., they are deposited relatively rapidly), that there are substantial spatial and temporal variations in both their concentrations and their composition, and that their contribution to acid dry deposition should not be ignored.

To evaluate deposition rates, several different approaches are possible. Field experiments can be conducted to monitor changes in some system of receptors from which average deposition rates can be deduced. More intensive experiments can measure the deposition of particular pollutants in some circumstances. Neither approach is capable of monitoring the long-term, spatial-average dry deposition of pollutants. To understand why, we must first consider in some detail the processes that influence pollutant fluxes and then relate these considerations to measurement and modeling techniques that are currently being advocated. The logical sequence illustrated in Figure C.2-1 will be used to guide this discussion.

2.1.2 Aerodynamic Factors

Except for the obvious difference that particles will settle slowly under the influence of gravity, small particles and trace gases behave similarly in the air. Trace gases are an integral part of the gas mixture that constitutes air and thus will be moved with all the turbulent motions that normally transport heat, momentum, and water vapor. However, particles have finite inertia and can fail to respond to rapid turbulent fluctuations. Table C.2-1 lists some relevant characteristics of spherical particles in air (based on data tabulated by Davies 1966, Friedlander 1977, and Fuchs 1964). The time scales of most turbulent motions in the air are considerably greater than the inertial relaxation (or stopping) times listed in the table. These time scales vary with height, but even as close as 1 cm from a smooth, flat surface, most turbulence energy will be associated with time scales longer that 0.01 sec, so that even 100-μm-diameter particles would follow most turbulent

TABLE C.2-1 Dynamic Characteristics of Unit Density Aerosol Particles at STP, Corrected for Stokes-Cunningham Effects[a]

Particle Radius (μm)	Diffusivity (cm^2/s)	Stopping Time (s)	Settling Speed (cm/s)
0.001	1.28×10^{-2}	1.33×10^{-9}	1.30×10^{-6}
0.007	3.23×10^{-3}	2.67×10^{-9}	2.62×10^{-6}
0.005	5.24×10^{-4}	6.76×10^{-9}	6.62×1^{-6}
0.01	1.35×10^{-4}	1.40×10^{-8}	1.37×10^{-5}
0.02	3.59×10^{-5}	2.97×10^{-8}	2.91×10^{-5}
0.05	6.82×10^{-6}	8.81×10^{-8}	8.63×10^{-5}
0.1	2.21×10^{-6}	2.28×10^{-7}	2.23×10^{-4}
0.2	8.32×10^{-7}	6.87×10^{-7}	6.73×10^{-4}
0.5	2.74×10^{-7}	3.54×10^{-6}	3.47×10^{-3}
1.0	1.27×10^{-7}	1.31×10^{-5}	1.28×10^{-2}
2.0	6.10×10^{-8}	5.03×10^{-5}	4.93×10^{-2}
5.0	2.38×10^{-8}	3.08×10^{-4}	3.02×10^{-1}
10.0	1.38×10^{-8}	1.23×10^{-3}	1.2×10^{0}

[a]Data are from Fuchs (1964), Davies (1966), and Friedlander (1977).

fluctuations. However, natural surfaces are normally neither smooth nor flat, and it is clear that in many circumstances the flux of particles will be limited by their inability to respond to rapid air motions.

Naturally occurring aerosol particles are not always spherical, although it seems reasonable to assume so in the case of hygroscopic particles in the submicrometer size range. Chamberlain (1975) documents the ratio of the terminal velocity of nonspherical particles to that of spherical particles with the same volume. In all cases, the nonspherical particles have a lower terminal settling speed than equivalent spheres. The settling speed differential is indicated by a "dynamical shape factor," α, as listed in Table C.2-2.

Thus, trace gases and small particles are carried by atmospheric turbulence as if they were integral components of the air itself, whereas large particles are also affected by gravitational settling, which causes them to fall through the turbulent eddies. In general, however, the distribution of pollutants in the lower atmosphere is governed by the dynamic structure of the atmosphere as much as by pollutant properties.

TABLE C.2-2 Dynamic Shape Factors as by which Nonspherical Particles Fall More Slowly than Spherical (from Chamberlain, 1975)

Shape	Ratio of axes	α
Ellipsoid	4	1.28
Cylinder	1	1.06
Cylinder	2	1.14
Cylinder	3	1.24
Cylinder	4	1.32
Two spheres touching, vertically	2	1.10
Two spheres touching, horizontally	2	1.17
Three spheres touching, as triangle	-	1.20
Three spheres touching, in line	3	1.34-1.40
Four spheres touching, in line	4	1.56-1.58

In daytime, the lower atmosphere is usually well mixed up to a height typically in the range 1 to 2 km, as a consequence of convection associated with surface heating by insolation. Pollutants residing anywhere within this mixed layer are effectively available for deposition through the many possible mechanisms. However, at night, the lower atmosphere becomes stably stratified and vertical transfer of nonsedimenting material is so slow that, at times, pollutants at heights as low as 50 to 100 m are isolated from surface deposition processes. Thus, in daytime, atmospheric transfer does not usually limit the rate of delivery of pollutants to the surface boundary layer in which direct deposition processes are active.

The fine details of turbulent transport of pollutants remain somewhat contentious. Notable among the areas of disagreement is the question of flux-gradient relationships in the surface boundary layer. It is now well accepted that the eddy diffusivity of sensible heat and water vapor exceeds that for momentum in unstable (i.e., daytime) but not in stable conditions over fairly smooth surfaces (see Dyer 1974, for example). However, it is not clear that the well-accepted relations governing either heat or momentum transfer are fully applicable to the case of particles or trace gases; some disagreement exists even in the case of water vapor. The situation is

even more uncertain in circumstances other than over large expanses of horizontally uniform pasture. When vegetation is tall, pollutant sinks are distributed throughout the canopy so that close similarity with the transfer of more familiar quantities such as heat or momentum is effectively lost. There is even considerable uncertainty about how to interpret profiles of temperature, humidity, and velocity above forests (see Garratt 1978, Hicks et al. 1979, Raupach et al. 1979).

2.1.3 The Quasi-laminar Layer

In the immediate vicinity of any receptor surface, a number of factors associated with the molecular diffusivity and the inertia of pollutants become important. Large particles carried by turbulence can be impacted on the surface as they fail to respond to rapid velocity changes. The physics of this process is similar to the physics of sampling by inertial collection.

Inertial impaction is a process that augments gravitational settling for particles that fall into a size range typically between 2- and 20-μm diameter (q.v. Slinn 1976b). Larger-sized particles tend to bounce, and capture is therefore less efficient, while smaller-sized particles experience difficulty in penetrating the quasi-laminar layer that envelops receptor surfaces. From the viewpoint of acidic particles, inertial impaction is a process of questionable relevance since most acidic species are associated with smaller particles (see Figure C.2-2), which are not strongly affected by this process. However, Figures C.2-2 and C.2-3 show that many airborne materials exist in the size range likely to be affected by inertial impaction. Since many of the chemical constituents of soil-derived particles are capable of neutralizing deposited acids, inertial impaction may have important indirect effects on acidic deposition.

To illustrate the role of molecular or Brownian diffusivity, it is informative to consider the simple case of a knife-edged thin plate, mounted horizontally and with edge normal to the wind sector. As air passes over (and under) the plate, a laminar layer develops, of thickness $\delta = c(vx/u)^{1/2}$, where v is kinematic viscocity, x is the downwind distance from the edge of the plate, and u is wind speed. According to Batchelor (1967), the value of the numerical constant c is 1.72. Thus, for a plate of dimensions 5 cm in a wind speed of

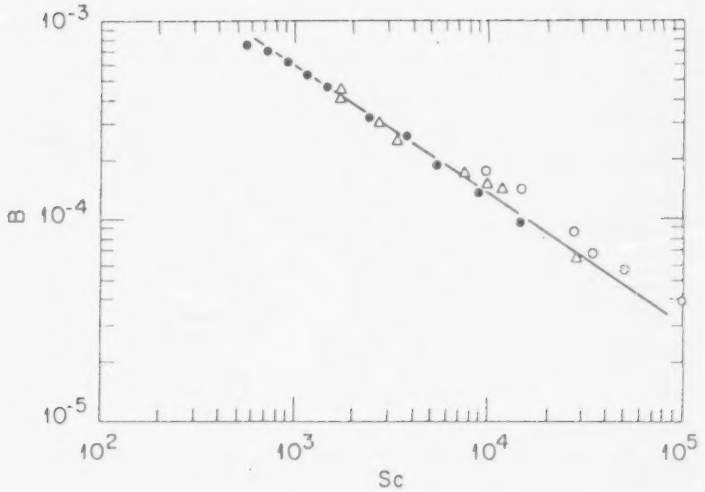

FIGURE C.2-3 Laboratory verification of Schmidt-number scaling for particle transfer to a smooth surface. The quantity plotted is $B \cong v_d/u_*$, evaluated for transfer across a quasi-laminar layer of molecular control immediately adjacent to a smooth surface. Data are from Harriott and Hamilton (1965; open circles), Hubbard and Lightfood (1966; triangles), and Muzushinz et al. (1971; solid circles), as reported by Lewellen and Sheng (1980). The line drawn through the data is Equation (C.2-1), with exponent $\alpha = -2/3$ and constant of proportionality $A \cong 0.06$.

1 m/s, we should imagine a boundary-layer thickness reaching about 1.5-mm thick at the trailing edge.

Over nonideal surfaces, the internal viscous boundary layer is frequently neither laminar nor constant with time. The layer generates slowly as a consequence of viscosity and surface drag as air moves across a surface. The Reynolds number Re ($\equiv ux/v$, where u is the wind speed, x is the downwind dimension of the obstacle, and v is kinematic viscosity) is an index of the likelihood that a truly laminar layer will occur. For large Re, air adjacent to the surface remains turbulent: viscosity is then incapable of exerting its influence. In many cases, it seems that the surface layer is intermittently turbulent. For these reasons, and because close similarlity between ideal surfaces studied in wind tunnels and natural surfaces is rather difficult to swallow, the term "quasi-laminar layer" is preferred.

Wind-tunnel studies of the transfer of particles to the walls of pipes, tend to support the concept of a limiting diffusive layer adjacent to smooth receptor

surfaces. Transfer across such a laminar layer is conveniently formulated in terms of the Schmidt number, Sc = v/D, where v is viscosity and D is the pollutant diffusivity. The conductance, or transfer velocity v_l, across the quasi-laminar layer is proportional to the friction velocity u_*:

$$v_l = Au_* Sc^{\alpha}, \qquad (C.2\text{-}1)$$

where A and α are determined experimentally. Most studies agree that the exponent α is about -2/3, as is evident in the experimental data represented in Figure C.2-3. However, a survey by Brutsaert (1975a) indicates exponents ranging from -0.4 to -0.8. The value of the constant A is also uncertain. The line drawn through the data of Figure C.2-3 corresponds to A ≈ 0.06, yet the wind-water tunnel results of Moller and Schumann (1970) appears to require A ≈ 0.6. These values span the value of A ≈ 0.2 recommended for the case of sulfur dioxide flux to fibrous, vegetated surfaces (Shepherd 1974, Wesely and Hicks 1977).

Laminar boundary-layer theory imposes the expectation that particle deposition to exposed surfaces will be strongly influenced by the size of the particle, with smaller particles being more readily deposited by diffusion than larger. It is clear that many artificial surfaces or structures made of mineral material will have characteristics for which the laminar-layer theories might be quite appropriate. However, the relevance to vegetation can be questioned. Microscale surface roughness elements can penetrate the barrier presented by this quasi-laminar layer and should be suspected as sites for enhanced deposition of both particles and gases (see Chamberlain 1980).

2.1.4 Phoretic Effects and Stefan Flow

Particles near a hot surface experience a force that tends to drive them away from the surface. For very small particles (<0.03-μm diameter, according to Davies 1967), this "thermophoresis" can be visualized as the consequence of hotter, more energetic air molecules impacting the side of the particle facing the hot surface. For larger particles, radiometric forces become important (Cadle 1966). In theory, thermal radiation can

cause temperature gradients across particles that are not good thermal conductors, resulting in a mean motion of the particle away from a hot surface. In summary, the thermophoresis depends on the local temperature gradient in the air, on the thermal properties of the particle, on the Krudsen number $Kn \equiv \lambda/r$ (where λ is the mean free path of air molecules and r is the radius of the particle), and on the nature of the interaction between the particle and air molecules (see Derjaguin and Yalamov 1972). As a rule of thumb, the thermophoretic velocity of very small particles (<0.03-μm diameter) is likely to be about 0.03 cm/s (estimated from values quoted by Davies 1967). For particles exceeding 1-μm diameter, the velocity will be about four times less.

The process of diffusiophoresis results when particles reside in a mixture of intermixing gases. In most natural circumstances, the principal concern is with water vapor. Close to an evaporating surface, a particle will be impacted by more water molecules on the nearer side. Since these water molecules are lighter than air molecules, there will be a net "diffusiophoresis" toward the evaporating surface. In essence, these "phoretic" forces result from the flow of molecules of some special kind through the gas mixture and the "drag" exerted on particles. Since diffusiophoresis and thermophoresis depend on the size and shape of the particle of interest, neither can be predicted with precision, nor can safe generalizations be made. These subjects are sufficiently complicated that they constitute specialities in their own right. Excellent discussions have been given by Friedlander (1977) and Twomey (1977). These phoretic forces vary with particle size but are generally small, and their influence on dry deposition can usually be disregarded.

Many workers include Stefan flow in general discussion of diffusiophoresis, but because of the conceptual difference between the mechanisms involved it seems better to consider them separately. Stefan flow results from injection into the gaseous medium of new gas molecules at an evaporating or subliming surface. Every gram-molecule of substrate material that becomes a gas displaces 22.41 liters of air, at STP. Thus, for example, a Stefan flow velocity of 22.41 mm/s will result when 18 g of water evaporates from a 1-m^2 area every second. Generalization to other temperatures and pressures is straightforward. Daytime evaporation rates from natural

vegetation often exceed 0.2 g/m^{-2} s for considerable times during the midday period, resulting in Stefan flow of more than 0.2 mm/s away from the surface. Detailed calculation for specific circumstances is quite simple. For the present, it is sufficient to note that Stefan flow is capable of modifying surface deposition rates by an amount that is larger than the deposition velocity appropriate for many small particles to aero- dynamically smooth surfaces.

Electrical forces have often been mentioned as possible mechanisms for promoting deposition (as well as the retention, see Section 2.1.5) of small particles, particularly through the "viscous" quasi-laminar layer immediately above receptor surfaces. Wason et al. (1973) report exceedingly high rates of deposition of particles in the size range 0.6 to 6 μm to the walls of pipes whenever a space charge is present. Chamberlain (1960) demonstrated the importance of electrostatic forces in modifying deposition velocities of small particles, when fields are sufficiently high. Plates charged to produce local field strengths of more than 2000 V cm^{-1} experienced considerably more deposition of small particles than uncharged plates, by factors between 2 and 15. However in fair-weather conditions, field strengths are typically less than 10 V cm^{-1} so that the net effect on particle transfer is likely to be small. Further studies of the ability of electrostatic forces to assist the transfer of particulate pollutants to vegetative surfaces were conducted by Langer (1965) and Rosinski and Nagomoto (1965). According to Hidy (1973), a series of experiments was conducted using single conifer needles and conifer trees. "For single needles or leaves, electrical charges on 2-μm-diameter ZnS dust with up to eight units of charge had no detectable effect at wind speeds of 1.2 to 1.6 m/s. The average collection efficiency was found to be 6% for edgewise cedar or fir needles, with broadside values an order of magnitude lower. Bounce-off after striking the collector was not detected, but re-entrainment could take place above 2 m/s wind speed. Tests on branches of cedar and fir by Rosinski and Nagomoto (1965) suggested similar results as for single needles." It should be noted, however, that the electrical mobility of a particle is a strong negative function of particle size, ranging from 2 cm/s per V/cm of field strength for 0.001-μm-diameter particles, to 0.0003 cm/s per V/cm for 0.1-μm particles (Davies 1967).

2.1.5 Surface Adhesion

Most workers assume that pollutants that contact a surface will be captured by it. For some gases, this assumption is clearly adequate. For example, nitric acid vapor is sufficiently reactive that most surfaces should act as nearly perfect sinks. Less reactive chemicals will be less efficiently captured. The case of particles is of special interest, however, because of the possibility of bounce and resuspension.

The role of electrostatic attraction in binding deposited particles to substrate surfaces remains something of a mystery. The process by which particles become charged and set up mirror-charges on the underlying surface is fairly well accepted. The resulting van der Waals forces are often mentioned as the major mechanism for binding particles once deposited. For large, nonspherical particles, dipole moments can be set up in natural electric fields, and these can help promote the adhesion at surfaces. These matters have been conveniently summarized by Billings and Gussman (1976), who provide mathematical relationships for evaluating the electrical energy of a particle on the basis of its size, shape, dielectric constant, and the strength of the surrounding electrical field. For smaller particles, the principal charging mechanism is thermal diffusion, leading to a Boltzmann charge distribution.

Condensation of water reduces the effectiveness of electrostatic adhesion forces, since leakage paths are then set up and charge differentials are diminished. However, the presence of liquid films at the interfaces between particles and surfaces causes a capillary adhesive force that compensates for the loss of electrostatic attraction. These "liquid-bridge" forces are most effective in high humidities and for coarse particles (<20 μm, according to Corn 1961).

Billings and Gussman (1976) draw attention to the effect of microscale surface roughness in promoting adhesion of particles to surfaces. Much of the experimental evidence is for particle diameters much greater than the height of surface irregularities (e.g., Bowden and Tabor 1950). It is the opposite case that is likely to be of greater interest in the present context, as will be discussed later.

2.1.6 Surface Biological Effects

The efficiency with which natural surfaces "capture" impacting particles or molecules will be influenced considerably by the chemical composition of the surface as well as its physical structure. The "lead candle" technique for detection of atmospheric sulfur dioxide is a historically interesting example of how chemical substrates can be selected to affect the deposition rates of particular pollutants.

Uptake rates of many trace gases by vegetation are controlled by biological factors such as stomatal resistance. In daytime, this is known to be the case for sulfur dioxide (Shepherd 1974, Spedding 1969, Wesely and Hicks 1977) and usually for ozone in most situations (Wesely et al. 1978). The similarity between sulfur and ozone is not complete, however, because the presence of liquid water on the foliage will tend to promote SO_2 deposition and to impede uptake of ozone; the former gas is quite soluble until the solution becomes too acidic, whereas the latter is essentially insoluble (q.v. Brimblecombe 1978).

Pubescence of leaves has received considerable attention. Chamberlain (1967) tested the roles of leaf stickiness and hairiness in a series of wind-tunnel tests. He concludes that "with the large particles (32 and 19 μm) the velocity of deposition to the sticky artificial grass was greater than to the real grass, but with those of 5 μm and less, it was the other way round, thus confirming . . . that hairiness is more important than stickiness for the capture of the smaller particles." The importance of leaf hairs appears to be verified by studies of the uptake of ^{210}Pb and ^{210}Po particles by tobacco leaves (Fleischer and Parungo 1974, Martell 1974) and by the wind-tunnel work of Wedding et al. (1975), who report increases by a factor of 10 in deposition rates for particles to pubescent leaves, compared with smooth, waxy leaves. It remains to be seen how greatly biological factors of this kind influence the rates of deposition of airborne particles to other kinds of vegetation.

2.1.7 Deposition to Liquid-Water Surfaces

Trace-gas and aerosol deposition on open water surfaces is of significant practical interest, especially consid-

ering the acidification of poorly buffered inland waters. Air blowing from land across a coastline will slowly equilibrate with the new surface at a rate that is strongly dependent on the stability regime involved. If the water is much warmer than upwind land, dynamic instability over the water will cause relatively rapid adjustment of the air to its new lower boundary, but if the water is cooler, stratified flow will occur and adjustment will be very slow. In the former (unstable) case, dry-deposition rates of all soluble or chemically reactive pollutants are likely to be much higher than in the latter. Clearly, air blowing over small lakes will be less likely to adjust to the water surface than when blowing over larger water bodies. Thus, during much of the summer, inland water surfaces will tend to be cooler than the air, and hence protected from dry deposition, because of the strongly stable stratification that will then prevail. This phenomenon will occur more frequently over small water bodies than larger ones (see Hess and Hicks 1975).

Following the guidance of chemical engineering gas-transfer studies, workers such as Kanwisher (1963), Liss (1973), and Liss and Slater (1974) have considered the role of Henry's law constant and chemical reactivity in controlling the rate of exchange of trace gases between the atmosphere and the ocean. In general, acidic and acidifying species like SO_2 are readily removed on contact with a water surface. Thus Hicks and Liss (1976) neglected liquid-phase resistance and derived net deposition velocities appropriate for the exchange of reactive gases across the air-sea interface. The work of Hicks and Liss is intended to apply to water bodies of sufficient size that the bulk exchange relationships of air-sea interaction research are applicable. Their considerations indicate that deposition velocities for highly soluble and chemically reactive gases such as NH_3, HCl, and SO_2 are likely to be between 0.10 and 0.15 percent of the wind speed measured at 10-m height. The analysis leading to this conclusion assumes that the molecular and eddy diffusivities can be combined by simple addition. This assumption has been shown to approximate the transfer of water vapor and sensible heat from water surfaces. However, for fluxes of trace gases the validity of this assumption is questionable. Slinn et al. (1978) argue that it is better to introduce molecular diffusivity through a term analogous to the Schmidt number of Equation (2-1), with the exponent

$\alpha = -2/3$. (In contrast, the linear assumption used by Hicks and Liss implies $\alpha = -1.0$.) In view of the uncertainties mentioned in discussion of Equation (C.2-1), further comment on the implications and ramifications of these alternative assumptions is not warranted.

In the limiting case of a trace gas of low solubility, the deposition velocity is determined by the large liquid-phase resistance, which is essentially proportional to the Henry's law constant.

It is probable that breaking waves will modify the simple gas-transfer formulations derived from chemical engineering pipe-flow and wind-tunnel work. It is not clear to what extent such features account for the apparent discrepancy between the various Schmidt number dependencies of the kind expressed by Equation (C.2-1). However, the fractional power laws are basically extensions of laboratory work, whereas the unit-power, additive-diffusivities result is an approximation to field data. It is to be hoped that the two approaches produce results that will converge in due course.

Figure C.2-4 previews the discussion of wind-tunnel particle deposition results that will be given later. Such wind-tunnel work indicates exceedingly low deposition velocities for particles in the size range of most acidic pollutants. As in the case of gas exchange, there are conceptual difficulties in extending these results to the open ocean. The role of waves in the transfer of small particles between the atmosphere and water surfaces remains essentially unknown. Not only does engulfment by breaking waves provide an alternative path across the quasi-laminar sublayer where molecular (or Brownian) diffusion normally controls the transfer, but also waves are a source of droplets that can scavenge particulate material from the air (see, however, the study of Alexander 1967, which indicates otherwise). Hicks and Williams (1979) have proposed a simple model of air-sea particle exchange that extends smooth-surface, wind- and water-tunnel results (as in Figure C.2-4) to natural circumstances by permitting rapid transfer to occur whenever waves break. This results in very low deposition velocities in light winds, but rapidly increasing when winds increase above about 5 m/s. Slinn and Slinn (1980) also suggest that particle transfer is more rapid than the wind-tunnel studies of Figure C.2-4 might indicate but present an alternative hypothesis for this more rapid transfer: that hygroscopic particles grow rapidly when exposed to high humidities such as are

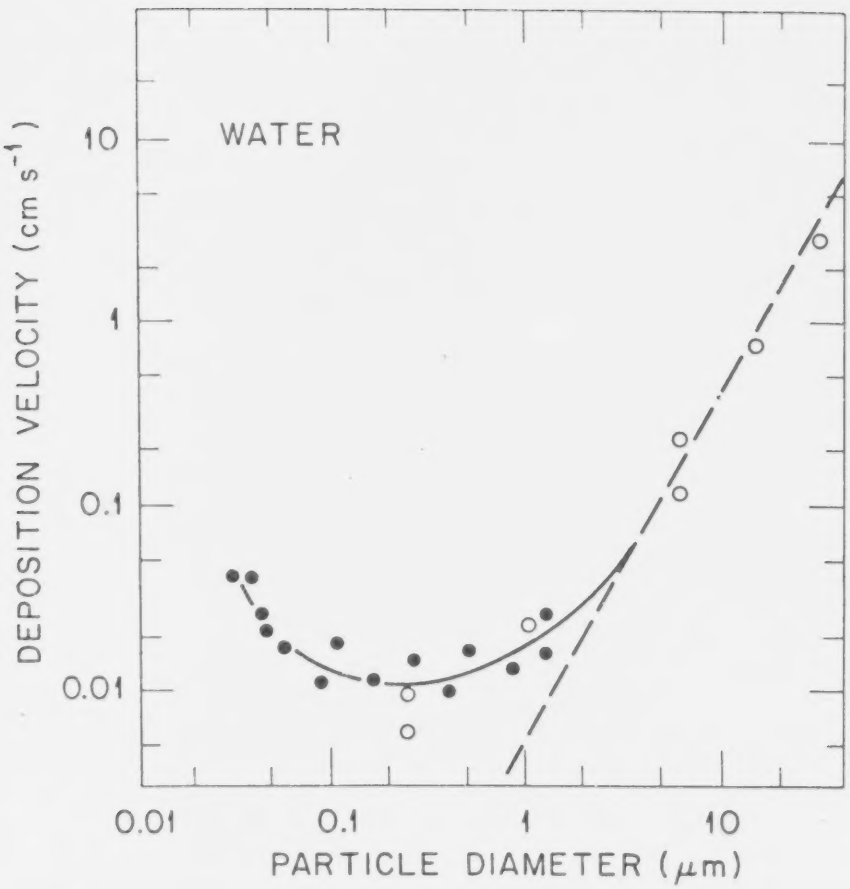

FIGURE C.2-4 Results of wind-tunnel studies of particle deposition to water surfaces. Solid circles are due to Moller and Schumann (1979), open circles to Sehmel and Sutter (1974). The dashed line at the right represents the terminal settling speed for 1.5 g cm^{-3} particles.

found in air adjacent to a water surface, resulting in increased gravitational settling and impaction to the water surface.

2.1.8 Deposition to Mineral Surfaces

Acid deposition is an obvious source of worry to architects, historians, and others concerned with the potentially accelerated deterioration of structures. Many popular building materials react chemically with acidic air pollutants, generating new chemical species

that can contribute directly to the decay process even if they are rapidly and efficiently washed off by precipitation. Furthermore, in some cases the chemical product causes a visual degradation that cannot easily be rectified, such as the blackening of metal work exposed to hydrogen sulfide.

The presence of water at the surface is known to be a key factor in promoting the fracturing and erosion of stone. Water penetrates pores and cracks and causes mechanical stresses both by freezing and by hydration and subsequent crystallization of salts (see Fassina 1978, Gauri 1978, Winkler and Wilhelm 1970). The earlier discussion of surface effects that influence dry deposition indicated that surface scratches and fractures will cause accelerated dry-deposition rates in localized areas. Moreover, phoretic effects are likely to be more important than in the case of foliage (because dry surfaces exhibit wider temperature extremes than moist vegetation). Stefan flow associated with dewfall is also probably more important than for vegetation. Hicks (1981) has summarized a number of relevant points as follows:

1. In daytime, particle fluxes will be greatest to the coolest parts of exposed surfaces.
2. Both particle and gas fluxes will be increased when condensation is taking place at the surface, and decreased when evaporation occurs.
3. If the surface is wet, impinging particles will have a better chance of adhering, and soluble trace gases will be more readily "captured."
4. The chemical nature of the surface is important; if reaction rates with deposited pollutants are rapid, then surfaces can act as nearly perfect sinks.
5. Biological factors can influence uptake rates, by modifying the ability of the surface to capture and bind pollutants.
6. The texture of the surface is important. Rough surfaces will provide better deposition substrates than smoother surfaces and will permit easier transport of pollutants across the near-surface quasi-laminar layer.

2.1.9. Fog and Dewfall

The processes that cause aerosol particles to nucleate, coalesce, and grow into cloud droplets are precisely the same as those that assist in the generation of fog.

Whenever surface air supersaturates, fog droplets form on whatever hygroscopic nuclei are available. These small droplets slowly settle onto exposed surfaces or are deposited by interception and impaction. The characteristics of the liquid that is deposited are much the same as those of cloud liquid water (see Section 3).

The conditions under which low-altitude surface fogs form are the cases of strong stratification in which vertical turbulent transport is minimized. The frequency of occurrence of fogs varies widely with location and with time of year. The depth is also highly variable. However, it must be assumed that fogs constitute a mechanism whereby the lower atmosphere (say the bottom hundred meters or so) can be cleansed of particulate and some gaseous pollutants.

At higher elevations, fog droplets are precisely the same as the cloud droplets that in other circumstances would grow and finally precipitate in substantially diluted form. The importance of cloud droplet interception has been demonstrated recently by Lovett et al. (1982) at an altitude of 1200 m in New Hampshire. Most of the net deposition of acidic species is by cloud droplet interception.

The presence of liquid water on exposed surfaces will obviously help promote the deposition of soluble gases and wettable particles. This surface water arises through the action of three separate mechanisms. Some plants expel fluid from foliage, usually at the tips of leaves, by a process known as guttation. Moisture can evaporate from the ground and recondense on other exposed surfaces, a mechanism known as distillation. However, these mechanisms are frequently confused with dewfall, which is properly the process by which water vapor condenses on surfaces directly from the air aloft. In practice, the origin of the surface moisture is immaterial to pollutants that come in contact with it. However, dewfall and distillation are processes that assist pollutant deposition through Stefan flow, whereas guttation does not. According to Monteith (1963), the maximum rate of dewfall is of the order of 0.07 mm/h, so that the maximum Stefan flow enhancement of the nocturnal deposition velocity is about 8 cm/h (see Section 2.1.4).

2.1.10 Resuspension and Surface Emission

Deposited particles can be resuspended into the air and subsequently redeposited. The mechanisms involved are much the same as those that cause saltation of particles from the beds of streams and from eroding croplands. These subjects are of great practical importance in their own right and have been studied at length. Concern about resuspension of radioactive particles near sites of accidents or weapons tests injected a note of some urgency into related studies during the 1950s and 1960s, as evidenced in the large number of papers on the subject included in the volume "Atmosphere-Surface Exchange of Particulate and Gaseous Pollutants" (Engelmann and Sehmel 1976).

The momentum transfer between the atmosphere and the surface is the driving force that causes surface particles to creep, bounce, and eventually saltate. There is a minimum frictional force that will cause particles of any particular size to rise from the surface. Bagnold (1954) identifies u_*^2 as a controlling parameter, so that it is the few occurrences of strongest winds that are the most important. While most thinking seems to center on widespread phenomena like dust storms, Sinclair (1976) points out that dust devils provide a highly efficient light-wind mechanism for resuspending surface particles and carrying them to considerable altitudes. Clearly, very large particles will not be moved frequently, or far. Very small particles are bound to the surface by adhesive forces that have already been discussed and tend to be protected in crevices or between larger particles.

Chamberlain (1982) has provided a theoretical basis for linking saltation of sand particles and snowflakes and for relating these phenomena to the generation of salt spray at sea.

It is not clear how saltation and related phenomena affect acid deposition. Surface particles that are injected into the air by the action of the wind do not normally move far, nor do they offer much opportunity for interaction with other air pollutants (firstly because they are confined in a fairly shallow layer near the suface and secondly because they have a very short residence time). Their effects are largely local.

Much smaller particles (in the submicrometer size range) are generated by reactions between atmospheric oxidants and organic trace gases emitted by some vegetation, especially conifers (see Arnts et al. 1978).

Once again, it is not obvious how these should best be considered in the present context of acid deposition. This is but one of many natural surface sources that provide a conceptual mechanism for injecting particles and trace gases into the lower atmosphere.

2.1.11 The Resistance Analog

Discussion of the relative importance of the various factors that contribute to the net flux of a particular atmospheric pollutant and determination of which process might be limiting in specific circumstances is simplified by considering a resistance model analogous to Ohm's law. Figure C.2-5 illustrates the way in which the concept is usually applied. An atmospheric resistance, r_a, is identified with the transfer of material through the air to the vicinity of the final receptor surfaces. This resistance is defined as that associated with the transfer of momentum; it is dependent on the roughness of the surface, the wind speed, and the prevailing atmospheric stability. The aerodynamic resistance can be written as

$$r_a = C_{fn}^{-1} - \psi_c/k)/u_*, \qquad \text{(C.2-2)}$$

where C_{fn} is the appropriate friction coefficient (the square root of the familiar drag coefficient) in neutral stability, u_* is the friction velocity (a scaling quantity defined as the root mean covariance between vertical and longitudinal wind fluctuations), k is the von Kármán constant, and ψ_c is a stability correction function that is positive in unstable, negative in stable, and zero in neutral stratifications (see Wesely and Hicks 1977). Equation (C.2-2) is obtained by straightforward manipulation of standard micrometeorological relations, as given by Wesely and Hicks, for example. The value of k is usually taken to be about 0.4. Table C.2-3 lists typical values of the friction coefficient for a range of surfaces.

The surface boundary resistance, r_b, is that which accounts for the difference between momentum transfer (i.e., frictional drag) at the surface and the passage of some particular pollutant through the near-surface quasi-laminar layer. In the agricultural meteorology literature, a quantity B^{-1} is frequently employed for this purpose (Brutsaert 1975a). The relationship between

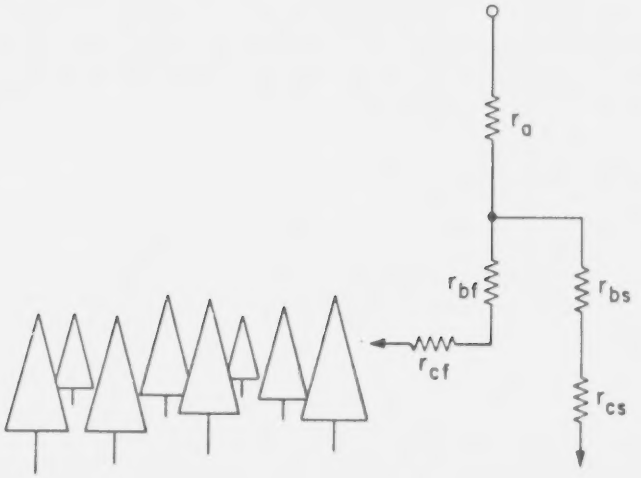

FIGURE C.2-5 A diagrammatic illustration of the resistance model frequently used to help formulate the roles of processes like those given in Figure C.2-1. Here, r_a is an aerodynamic resistance controlled by turbulence and strongly affected by atmospheric stability, r_{bf} and r_{bs} represent surface boundary-layer resistances that are determined by molecular diffusivity and surface roughness, and r_{cf} and r_{cs} are the net residual resistances required to quantify the overall deposition process, to the eventual sink. The second subscripts f and s are intended to indicate pathways to foliage and to soil, respectively. There are many other pathways that might be important; the diagram is not intended to be more than a simple visualization of some of the important factors.

these quantities can be clarified by relating both to the micrometeorological concept of a roughness length, z_o (the height of apparent origin of the neutral logarithmic wind profile). Then the total atmospheric resistance, R, between the surface in question and the height of measurement, z, can be written as

$$\begin{aligned} R &= (ku_*)^{-1}[\ln(z/z_{oc}) - \psi_c] \\ &= (ku_*)^{-1}[\ln(z/z_o) + \ln(z_o/z_{oc}) - \psi_c] \\ &= r_a + (ku_*)^{-1} \cdot \ln(z_o/z_{oc}), \end{aligned} \qquad \text{(C.2-3)}$$

where z_{oc} is a roughness length scale appropriate for the transfer of the pollutant. The residual boundary-layer resistance, $r_b = R - r_a$, is then

$$r_b = (ku_*)^{-1} \cdot \ln(z_o/z_{oc}), \qquad \text{(C.2-4)}$$

which alternatively is written as

$$r_b = (u_* B)^{-1}. \qquad \text{(C.2-5)}$$

B is, therefore, a measure of the nondimensionalized limiting deposition velocity for concentrations measured sufficiently close to a receptor surface that the resistance to momentum transfer can be disregarded.

It should be noted that some workers refer to r_b as the aerodynamic resistance and use the symbol r_a for it (e.g., O'Dell et al. 1977).

Shepherd (1974) recommends using a constant value $kB^{-1} = \ln(z_o/z_{oc}) = 2.0$ for transfer to vegetation, on the basis of results obtained over rough, vegetated surfaces. However, the role of the Schmidt number in accounting for diffusion near a surface needs to be taken into account. Wesely and Hicks (1977) advocate the use of a Schmidt number relationship like that of Equation (C.2-1), so that the surface boundary-layer resistance would then be written as

$$r_b = 5\ Sc^{2/3} u_*. \qquad \text{(C.2-6)}$$

Equation (C.2-6) implies a value of 0.2 for A in the boundary-layer relationship given by Equation (C.2-1), as was mentioned earlier.

The final resistances in the conceptual chain of processes represented diagrammatically by Figure C.2-5

TABLE C.2-3 Estimates of Roughness Characteristics Typical of Natural Surfaces[a]

Surface	Approx. Canopy Height (m)	Roughness Length (cm)	Neutral Friction Coefficient, C_{fn}
Smooth ice	0	0.003	0.042
Ocean	0	0.005	0.045
Sandy desert	0	0.03	0.055
Tilled soil	0	0.10	0.066
Thin grass	0.1	0.70	0.095
Tall thin grass	0.5	5.	0.16
Tall thick grass	0.5	10.	0.21
Shrubs	1.5	20.	0.25
Corn	2.3	30.	0.29
Forest	10.	50.	0.23
Forest	20.	100.	0.24

[a]Values of the friction coefficient $C_{fn} (\equiv u_*/u)$ are evaluated for neutral conditions, at a height 50 cm above the surface or top of the canopy.

are those that permit material to be transferred to the surface itself. For many pollutants, it is necessary only to consider the canopy foliage resistance r_{cf}, but for some it is also necessary to consider uptake at the ground by invoking a resistance to transfer to soil (or a forest floor), r_{cs}. In concept, it is also appropriate to differentiate between boundary-layer resistances r_{bf} and r_{bs}, for transfer to foliage and soil, respectively, as is shown in the diagram. Many other resistances can be identified and might often need to be considered, but further complication of Figure C.2-5 is unnecessary. Its main purpose is illustrative.

Transfer of many trace gases to foliage occurs by way of stomatal uptake, which, because of stomatal physiology, imposes a strong diurnal cycle on the overall deposition behavior. Following initial work by Spedding (1969), studies of foliar uptake of sulfur dioxide have repeatedly confirmed the controlling role of stomatal resistance. Chamberlain (1980) summarizes results of experiments by Belot (1975) and Garland and Branson (1977), who compared surface conductances of sulfur dioxide with those for water vapor over a broad range of stomatal openings (which largely govern stomatal resistance). The conclusion that stomatal resistance is the controlling factor when stomates are open appears to be well founded. However, once again, it is necessary to apply corrections to account for the diffusivity of the trace gas in question; the higher the molecular diffusivity of the gas, the lower the stomatal resistance.

Fowler and Unsworth (1979) point out that SO_2 deposition to wheat continues, even when stomates are closed, at a rate that suggests significant deposition at the leaf cuticle. Thus, it is not always sufficient to compute the canopy-foliage resistance r_{cf} on the assumption that SO_2 uptake is via stomates alone (although this may indeed be a sufficient approximation in most circumstances). Instead, it is more realistic to estimate r_{cf} from its component parts via

$$r_{cf} \approx (r_{st}^{-1} + r_{cut}^{-1})^{-1}/(LAI) \qquad (C.2\text{-}7)$$

(following Chamberlain 1980), where r_{st} is the stomatal resistance, and r_{cut} is cuticular resistance. LAI is the leaf area index (the total area of foliage per unit horizontal surface area). Note that in most literature the LAI is assumed to be the single-sided leaf area index. However, sometimes both sides of the leaves are counted.

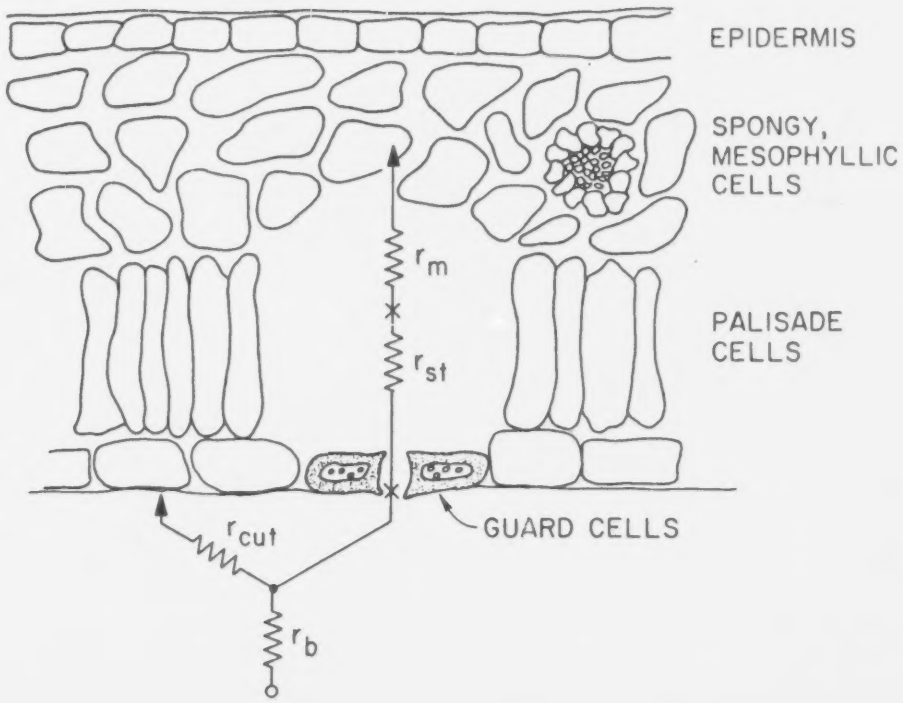

FIGURE C.2-6 An illustration of the roles of different resistances associated with trace gases uptake by a leaf. Material is transferred along several possible pathways, of which two are shown. These involve cuticular uptake via a resistance, r_{cut}, and transfer through stomatal pores (via r_{st}) into substomatal cavities, with subsequent transfer to mesophyllic tissue (via r_m). The way in which the various resistances are combined to provide the best visualization of the overall transfer process is not clear.

The resistance analogy permits a closer look at the mechanisms that transfer gaseous material to leaves. Figure C.2-6 illustrates the pathways involved: via stomatal openings into the interior of the leaf (involving stomatal and mesophyll resistances, r_{st} and r_m) or through the epidermis (involving a cuticular resistance, r_{cut}).

The resistance model is somewhat limited by the manner in which it structures the chain of relevant processes, each being represented by a resistance to transfer that occupies a prescribed location in a conceptual network. The structure of this network is sometimes not clear, and furthermore, there are important processes that do not conveniently fit into the resistance model. Mean drift velocities (e.g., gravitational settling of particles) are not easily accommodated in the simple resistance

picture, and it is doubtful whether some of the biological factors are relevant to the question of particle transfer. Studies of leaves show that stomates are typically slits of the order of 2-20 μm long. For stomatal uptake of particles to be a controlling factor of deposition, we would need to hypothesize spectacularly good aim by the particles.

2.2 METHODS FOR MEASURING DRY DEPOSITION

2.2.1 Direct Measurement

There is little question that the deposition of large particles is accurately measured by collection devices exposed carefully above a surface of interest. Deposit gauges and dust buckets have been important weapons in the geochemical armory for a long time. They are intended to measure the rate of deposition of particles that are sufficiently large that deposition is controlled by gravity. In studies of radioactive fallout conducted in the 1950s and 1960s, these same devices were used. In the case of debris from weapons tests, the major local fallout was of so-called hot particles, originating with the fragmentation of the casing of the weapon and its supporting structures, and the suspension of soil in the vicinity of the explosion. These large particles fall over an area of rather limited extent downwind of the explosion. This area of greatest fallout was the major focus of the work on fallout dry deposition. It was largely in this context that dustfall buckets were used to obtain an estimate of how much radioactive deposition occurred. It was recognized that collection vessels failed to reproduce the microscale roughness features of natural surfaces. However, this was not seen as a major problem, since the emphasis was on evaluating the maximum rate of deposition that was likely to occur, so that upper limits could be placed on the extent of possible hazards. Nevertheless, efforts were made to "calibrate" collection vessels in terms of fluxes to specific types of vegetation, soils, etc. (see Hardy and Harley 1958).

Much further downwind, most of the deposition was shown to be associated with precipitation, since the effective source of the radioactive fallout being deposited was typically in the upper troposphere or the lower stratosphere. The acknowledged inadequacies of collection buckets for dry deposition were then of only

little concern, since dry fallout composed a small fraction of the total surface flux.

In the context of present concerns about acid deposition, we must worry not only about large, gravitationally settling particles but also about small "accumulation-size-range" particles that are formed in the air from gaseous precursors and about trace gases themselves. All of these materials contribute to the net flux of acidic and acidifying substances by dry processes. It is known that collection vessels do indeed provide a measure of the flux of large particles. However, accumulation-size-range particles, typically of less than 1-μm diameter, do not deposit by gravitational settling at a significant rate. These small particles are transported by turbulence through the lower atmosphere and are deposited by impaction and interception on surface roughness elements, with the assistance of a wide range of surface-related effects (e.g., electrophoresis, Stefan flow) many of which will be influenced by the detailed structure of the surface involved.

Early work on the deposition of radioactive fallout made use of collection vessels and surrogate surface techniques that were frequently "calibrated" in terms of fluxes to specific types of vegetation, soils, etc. Studies of this kind were relatively easy, especially in the case of radioactive pollutants, since very small quantities of many important species could be measured accurately by straightforward techniques. Most of the radioactive materials that were of interest do not exist in nature, and so experimental studies benefitted from a zero background against which to compare observed data. Moreover, major emphasis was on the dose of radioactivity to specific receptors, a quantity that is strongly influenced by contributions of large, "hot" particles in situations of practical interest. Such circumstances included deposition of bomb debris, fission products, and soil particles from the radioactive cloud downwind of nuclear explosions. In such cases, highest doses were incurred near the source and were due to these larger particles.

The applicability of collection vessels and surrogate surfaces in studies of the dry deposition of acidic pollutants is in dispute. Principal among the conceptual difficulties concerning their use is their inability to reproduce the detailed physical, chemical, and biological characteristics of natural surfaces, which are known to control (or at least strongly influence) pollutant uptake

in most instances. Furthermore, the continued exposure of already-deposited materials to airborne trace gases and aerosol particles undoubtedly causes some changes to occur, but of unpredictable magnitude and unknown significance. A recent intercomparison between different kinds of surrogate surfaces and collection vessels has indicated that fluxes derived from exposing dry buckets are more than those obtained using small dishes, which in turn exceed values obtained using rimless flat plates (Dolske and Gatz 1982). This provides a tantalizing tidbit of evidence for an ordering of performance characteristics according to the total exposed surface area per unit horizontal projection. In this context, the similarity with arguments concerning leaf area index seems especially attractive. Micrometeorological data obtained during the same experiment fall between the extremes represented by the buckets and the flat plates.

Dasch (1982) reports on a comparison between many different configurations of flat-plate collection surfaces, pans, and buckets. The results indicate that glass surfaces provide the greatest flux estimates for almost all chemical species considered and Teflon the lowest. Bucket data generally fall midway in the range.

Tracer techniques that were developed in the radioecology era for investigating fluxes to natural surfaces offer some promise. A β-emitting isotope of sulfur, ^{35}S, lends itself to use in studies of SO_2 uptake by crops since measurements of low rates of sulfur accumulation are then possible. Garland (1977), Garland and Branson (1977), Garland et al. (1973), and Owers and Powell (1974) report the results of a number of studies of $^{35}SO_2$ uptake by various vegetated surfaces ranging from pasture to pine plantation and by nonvegetated surfaces such as water.

In concept, it is feasible to extend studies of this kind to the deposition of sulfurous particles, but as yet no such experiment has been reported. However, analogous studies of particle deposition using nonradioactive aerosol tracers have been carried out. In wind-tunnel experiments, Wedding et al. (1975) employed uranine dye particles in conjunction with lead chloride particles to study the influence of leaf microscale roughness on particle capture characteristics; uranine particles are relatively easily measured by fluorimetry, whereas measurements of lead deposition require far more painstaking chemical analysis of the deposition surface. The particle sizes used by Wedding et al. were in the range of 3- to 7-μm diameter.

Considerably larger particles have been used in many studies. In detailed wind-tunnel studies, Chamberlain (1967) used lycopodium spores (~30-μm aerodynamic diameter). Workers at Brookhaven National Laboratory extended these wind-tunnel techniques to real-world circumstances by conducting a series of experiments employing pollen grains in the same general size range (Raynor et al. 1970, 1971, 1972, 1974).

In general, these methods of tracer measurement have not been applied to natural circumstances for the particle sizes of major interest in the present context of acid deposition. An important exception concerns studies of deposition on snow surfaces. The retention of deposited material at the top of or within a snowpack has been studied in some detail and continues to be an intriguing area of research. Particulate materials such as sulfate were considered by Dovland and Eliassen (1976), who studied the accumulation upon snow surfaces during periods of no precipitation and found average deposition velocities in the range 0.1 to 0.7 cm/s depending on the assumption made regarding the contribution by gaseous SO_2 deposition. Similar work by Barrie and Walmsley (1978) yielded average sulfur dioxide deposition velocities to snow in the range 0.3 to 0.4 cm/s, with standard error equivalent to about a factor or 2.

Dillon et al. (1982) and Eaton et al. (1978) present examples of the use of calibrated watersheds to estimate atmospheric deposition. Dry-deposition fluxes are estimated as a residual between measured fluxes out of a conceptually closed system and measured wet deposition into it. Considerable effort is required to document annual chemical mass balances for specific watersheds. Once the effort is made, it appears possible to draw fairly well-founded conclusions regarding dry deposition, although obviously such estimates might result as the difference between fairly large numbers. According to Eaton et al., the annual dry-deposition flux estimate obtained at the Hubbard Brook Experimental Forest in New Hampshire is accurate to about ±35 percent (one standard error). The data do not permit apportionment between gaseous and particulate sulfur inputs, but the total sulfur flux corresponded to a deposition velocity of about 0.6 cm/s.

2.2.2 Laboratory Studies

Figure C.2-1 illustrates the overall complexity of the problem of dry deposition. While it is indisputable that no indoor experiment can provide a comprehensive evaluation of pollutant deposition that would be applicable to the natural countryside, laboratory studies provide the unique attraction of controllable conditions. It is feasible to study the relative importance of various factors thought to be of importance, as in Figure C.2-1 and especially as in Figure C.2-6, and to formulate these processes in a logical manner. In this general category, we must include the extensive wind-tunnel work referred to earlier, the pipe-flow and flat-plate studies conducted in experiments more aligned to problems of chemical engineering, and the chamber experiments favored by ecologists and plant physiologists. Distinction between these kinds of experiment is often difficult. Many exposure chambers and pipe-flow studies have features of wind tunnels.

The utility of chamber studies is well illustrated by the series of results reported by Hill (1971). By comparing the rates of deposition of various trace gases to oat and alfalfa canopies exposed in large chambers, Hill concluded that solubility was a critical parameter in determining uptake rates of trace gases by vegetation. The ordering of deposition velocities was: hydrogen fluoride > sulfur dioxide > chlorine > nitrogen dioxide > ozone > carbon dioxide > nitric oxide > carbon monoxide. Furthermore, the chamber studies indicated a wind-speed dependence of the kind predicted by turbulent transfer theory and demonstrated a physiological effect of chlorine and ozone on stomatal opening: exposure to high concentrations of either quantity caused partial stomatal closure, thus limiting the fluxes of all trace gases that are stomatally controlled.

Experiments conducted by Judeikis and Wren (1977, 1978) yielded valuable information on the deposition of hydrogen sulfide, dimethyl sulfide, sulfur dicxide, nitric oxide, and nitrogen dioxide to nonvegetated surfaces (Table C.2-4). The values listed were derived from initial deposition rates obtained before surface accumulation limited uptake rates. For comparison, surface resistances derived from Hill's (1971) studies of trace-gas uptake by alfalfa are also listed. On the whole, the ordering of deposition velocities suggested by

Hill's work appears to be supported, providing some justification for extending the ordering to CO, H_2S, and $(CH_3)_2S$ in the manner indicated in the table. Residual surface resistance to uptake of soluble gases by solid, dry surfaces appears to be substantially greater than for vegetation, which is as would be expected.

The values listed in Table C.2-4 represent resistances to transport very near the surface, to which atmospheric resistances must be added to obtain values representative of natural, outdoor conditions. The reciprocals of the tabulated numbers provide upper limits of the appropriate deposition velocities.

Similarly informative data have been obtained about particle deposition on surfaces that can be contained in wind tunnels. Studies of this kind are an obvious extension of pipe-flow investigations by workers such as Friedlander and Johnstone (1957) and Liu and Agarwal (1974), which provide strong support for theories involving particle inertia and Schmidt number scaling. Wind tunnels provide a means to extend chamber and

TABLE C.2-4 Resistances to Deposition of Selected Trace Gases, Measured for Solid Surfaces in a Cylindrical Flow Reactor (Judeikis and Stewart, 1976) and for Alfalfa in a Growth Chamber (Hill, 1971); Solid-Surface Data are Derived from Table 2 of Judeikis and Wren (1978); the Alfalfa Values are Obtained from Table 1 of Hill (1971)

Pollutant	Substrate Surface		
	Adobe Clay	Sandy Loam	Alfalfa
CO	-	-	∞
H_2S	62	67	-
$(CH_3)_2S$	3.6	16	-
NO	7.7	5.3	10.
CO_2	-	-	3.3
O_3	-	-	0.7
NO_2	1.3	1.7	0.5
Cl_2	-	-	0.5
SO_2	1.1	1.7	0.4
HF	-	-	0.3

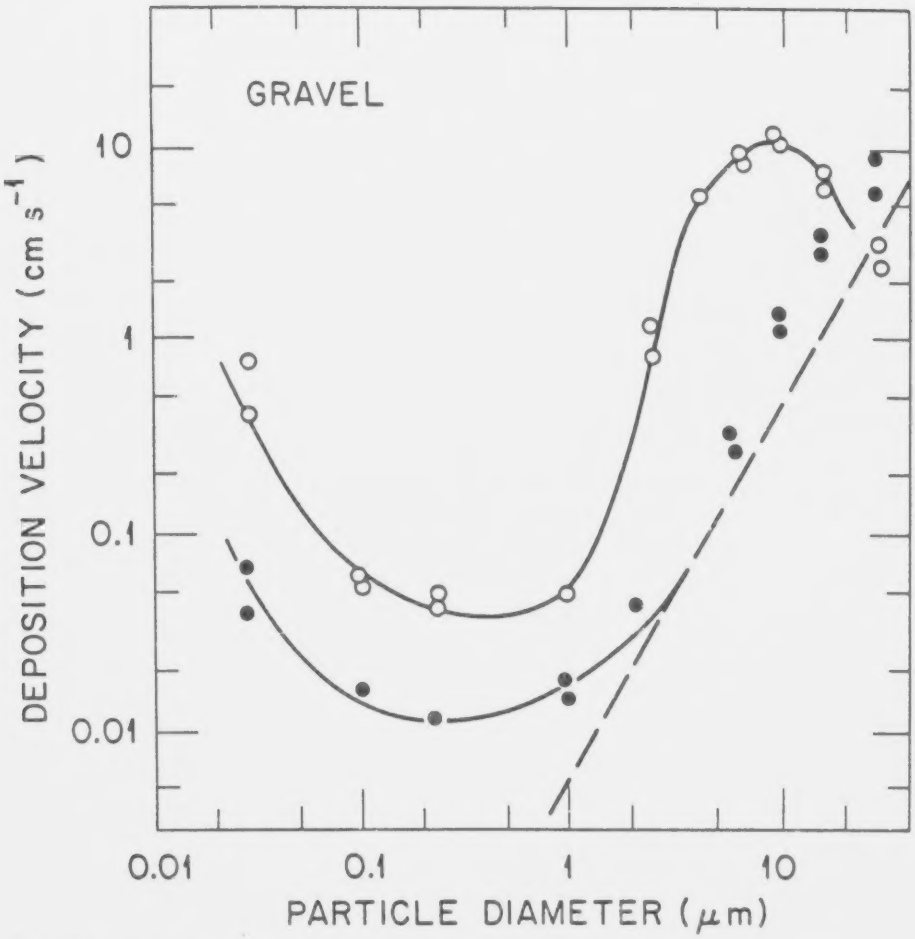

FIGURE C.2-7 Results of wind-tunnel studies of particle deposition to 1.6-cm-diameter dry, gravel (after Sehmel et al. 1973a). Solid circles were obtained at about 2.4 m/s, open circles at about 16 m/s.

pipe-flow investigations to situations more closely approximating natural conditions.

Results obtained in studies of particle deposition to dry gravel (Sehmel et al. 1973a) are shown in Figure C.2-7. The wind-speed effect evident in these data is fairly typical and applies also in the case of vegetation (Figure C.2-8). Experiments on deposition to wet gravel were also conducted. These indicated deposition velocities some 30 percent less than the values evident in Figure C.2-7 (for particles in the 0.2- to 1.0-μm

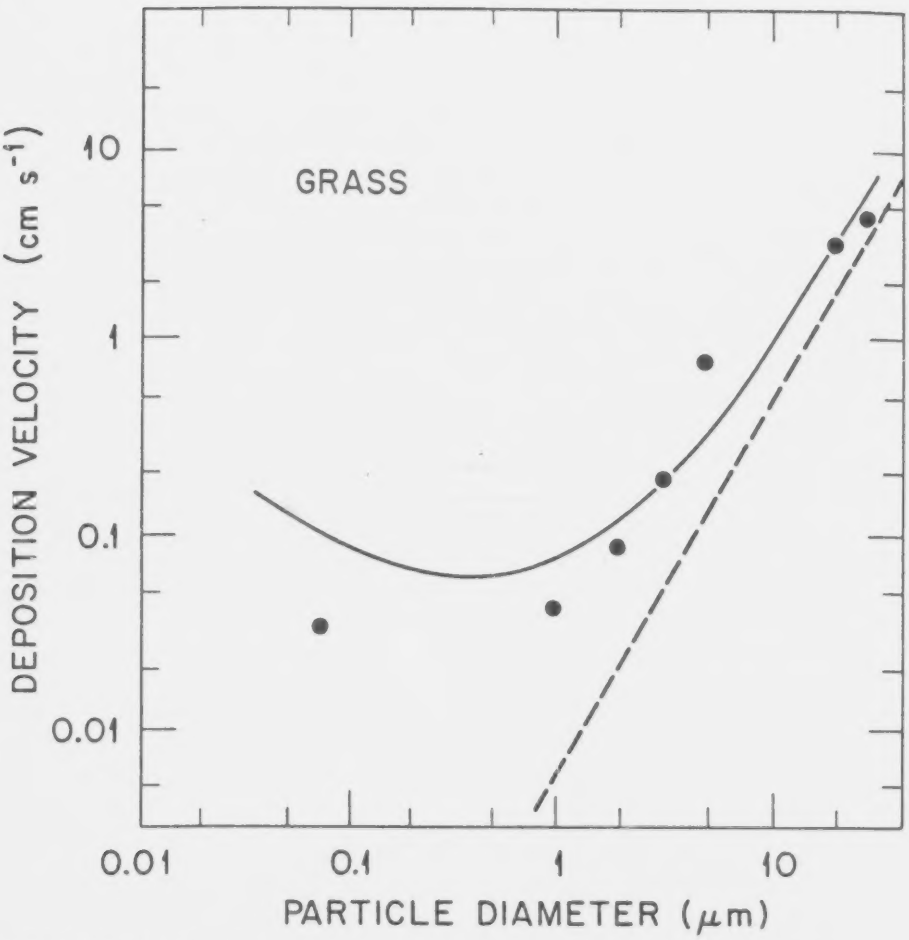

FIGURE C.2-8 Results of wind-tunnel studies of particle deposition to grass as reported by Chamberlain (1967; dots; natural grass; $u_* \cong 70$ cm/s) and by Sehmel et al. (1973; the curve; artificial grass; $u_* \cong 16$ cm/s).

size range), as might be expected from considerations of Stefan flow and diffusiophoresis. When surface roughness was increased, deposition velocities also increased.

Chamberlain (1967) extended his earlier (1966) wind-tunnel studies of gas transfer to "grass and grass-like surfaces" by considering particle deposition to rough surfaces. Sehmel (1970) conducted similar wind-tunnel experiments, employing monodisperse particles ranging from about 0.5- to 20-μm diameter. Figure C.2-8 combines results from Chamberlain (1967), and Sehmel et al. (1973b). The Chamberlain data refer to live grass, but the Sehmel et al. data were obtained

using 0.7-cm-high artificial grass. Moreover, the two sets of data were obtained at different wind speeds (Chamberlain, $u_* \approx 70$ cm/s; Sehmel et al., $u_* \approx 19$ cm/s). Further tests conducted by Chamberlain (1967) indicated that deposition velocities to natural grass exceeded those to artificial grass by a factor of about 2 for particles smaller than about 5 μm. This appears to be contrary to the indication of Figure C.2-7, where v_d (natural) of Chamberlain is seen to be about half the v_d (artificial) of Sehmel et al. for sizes less than a few micrometers. The difference in u_* between their experiments amplifies this discrepancy, rather than resolving it. However, both data sets provide evidence for the deposition velocity particle-size dependency that is predicted by theory and supported by all such laboratory investigations.

2.2.3 Micrometeorological Measurement Methods

The factors that control pollutant fluxes are frequently surface properties such as stomatal resistance and soil moisture (for soluble gases), cuticular resistance and the available leaf area (for strongly surface-reactive gases like HF and HNO_3), and microscale roughness (for submicrometer particles). Any measurement technique that interferes with a controlling property is likely to yield erroneous results, and hence there has been considerable effort expended to develop and apply methods of measurement that impose no surface or environmental modification. In concept, if an area is sufficiently homogeneous, flat, and contains no areas of strong sources or sinks, pollutant fluxes can be assumed to be constant with height. Therefore questions regarding dry deposition can be addressed by measuring the flux of material through a horizontal layer of air at some more convenient height above the surface. The intent of any such study is to investigate dry-deposition fluxes in carefully documented natural situations in order to identify and quantify controlling properties. The results of these investigations are formulations of surface mechanisms, surface boundary-layer resistances, stomatal resistances, etc. The demanding site criteria are required to enable these results to be obtained from the experiments; the surface parameterizations that are derived are far more widely applicable.

Several micrometeorological methods are suitable for measuring dry-deposition fluxes in intensive case studies. The flux can be measured directly by eddy correlation, a process that evaluates instantaneous products of the the vertical wind speed, w, and pollutant concentration, C, in order to derive the time-average vertical flux F_c as

$$F_c = \overline{\rho w' C'}, \qquad \text{(C.2-8)}$$

where ρ is air density and the primes denote deviations from mean values. The over-bar indicates a time average. This is an extremely demanding task and constitutes a specialized field of micrometeorology in its own right. Details of experimental procedures are given by, for example, Dyer and Maher (1965), Kaimal (1975), and Kanemasu et al. (1979).

Figure C.2-9 shows some examples of sensor output signals that are fundamental to the eddy-correlation technique. Fast-response sensors of any pollutant concentration can be used; the trace shown for CO_2 in the diagram is an interesting example of considerable agricultural relevance. As a basic requirement, sensors suitable for eddy-correlation applications should have response times shorter than 1 sec for operation at convenient heights on towers. For application aboard aircraft (Bean et al. 1972, Lenschow et al. 1980), considerably faster response is required.

Eddy-correlation methods have been used in field experiments addressing the fluxes of ozone (Eastman and Stedman 1977), sulfur (Galbally et al. 1979, Hicks and Wesely 1980), nitrogen oxides (Wesely et al. 1982b), carbon dioxide (Desjardins and Lemon 1974, Jones and Smith 1977), and small particles (Wesely et al. 1977).

Rates of transfer through the lower atmosphere are governed by intensities of turbulence generated by both mechanical mixing and convection. In this context, there are three atmospheric quantities that cannot be separated: the vertical flux of a material, the local gradient of its concentration ($\partial C/\partial z$), and its corresponding eddy diffusivity (K). Knowledge of any two of these quantities will permit the third to be evaluated. Often, when sensors suitable for direct measurement of pollutant fluxes are not available, assumptions regarding the eddy diffusivity are made to

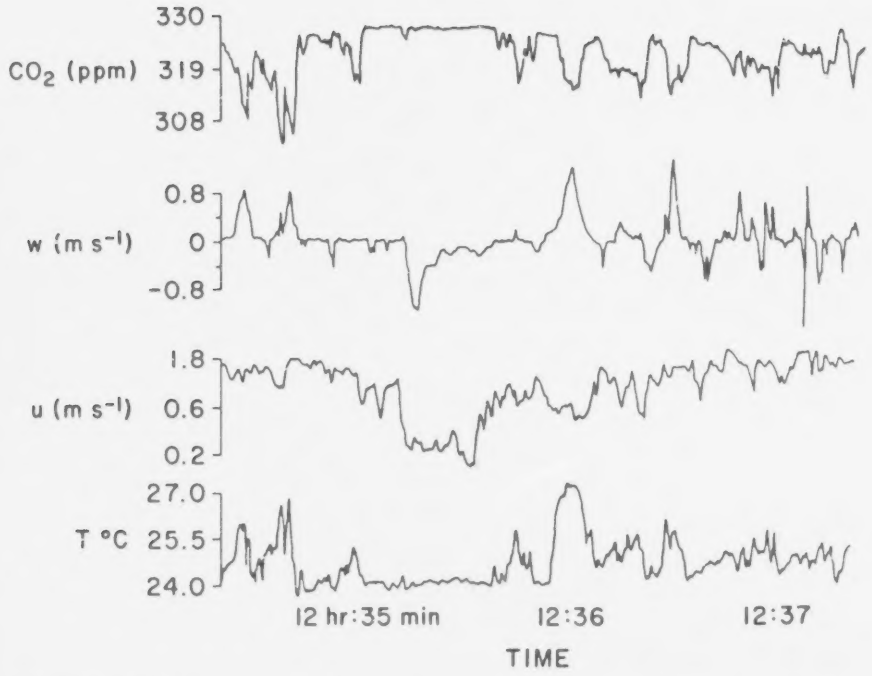

FIGURE C.2-9 An example of atmospheric turbulence near the surface. These traces of CO_2 concentration, vertical velocity (w), wind speed (u), and temperature (T) were obtained over a corn canopy by workers at Cornell University.

provide a method for estimating fluxes from measurements of vertical concentration gradients:

$$F_c = \rho K(\partial C/\partial z). \qquad (C.2\text{-}9)$$

Droppo (1980) and Hicks and Wesely (1978) have summarized a number of critical considerations. In particular, with a typical value of $u_* = 40$ cm/s and neutral stability, the concentration difference between adjacent levels differing in height by a factor of 2 is about 9 percent, for a 1 cm/s deposition velocity. In unstable (daytime) conditions, smaller gradients would be expected for the same v_d; in stable conditions, they would be greater.

The demands for high resolution by the concentration measurement technique are obvious. Nevertheless, a substantial quantity of excellent information has been obtained, especially concerning fluxes of SO_2 (Fowler 1978, Garland 1977, Whelpdale and Shaw 1974).

It should be emphasized that the stringent site uniformity requirements mentioned above for use of

eddy-correlation approaches are also relevant for gradient studies. The detection of a statistically significant difference between concentrations at two heights is not necessarily evidence of a vertical flux and can only be interpreted as such after the appropriate siting criteria have been satisfied.

Gradients of particle concentration present special problems since it is often not possible to derive internally consistent results from alternative measurements. Droppo (1980) concludes that "The particulate source and sink processes over natural surfaces cannot be considered as a simple unidirectional single-rate flux." Thus, the proper interpretation of gradient data in terms of fluxes might not be possible for airborne particles, even in the best of siting circumstances, because of the role of the surface in emitting and resuspending particles. In this case, eddy-correlation methods will still provide an accurate determination of the flux through a particular level, but this flux will be made up of a downward flux of airborne material and an upward flux of similar material of surface origin. Disentangling the two is likely to present a considerable problem.

None of the various micrometeorological methods has yet been developed to the extent necessary for routine application. Rather, they are research methods that can be used in specific circumstances, requiring considerable experimental care and the use of sensitive equipment and fairly complicated data analysis. They are more suitable for investigating the processes that control dry deposition than for monitoring the flux itself. Nevertheless, some new techniques that might be appropriate for dry-deposition monitoring are currently under development. A "modified Bowen ratio" method is being developed in the hope that it might permit an accurate determination of vertical fluxes without the need for rapid response or great resolution (Hicks et al. 1981). High-frequency variance methods are also being advocated but have yet to be fully investigated; for these, sensors having rapid response are required. An eddy-accumulation method that bypasses the need for rapid response of the pollutant sensor is of long-standing interest (e.g., Desjardins 1977) but has yet to be applied to the pollutant flux problem with significant success.

2.3 FIELD INVESTIGATIONS OF DRY DEPOSITION

2.3.1 Gaseous Pollutants

Table C.2-5 summarizes a number of recent field experiments on trace gas deposition. The listing is drawn from a variety of sources (especially Chamberlain 1980, Garland 1979, Sehmel 1979, 1980a); it is not meant to be exhaustive but is intended to demonstrate that much of the available data on surface fluxes of trace gases refers to daytime conditions, when "canopy" resistances are usually the controlling factors. Extrapolation of these values to nighttime conditions is dangerous on two grounds; first, because of the large changes that might accompany stomatal closure and, second, because of the much greater influence of aerodynamic resistance in nighttime, stable conditions.

Figure C.2-10 illustrates the large diurnal cycle that is typical of the dry-deposition rates of most pollutants. These observations were made over a pine plantation in North Carolina, using eddy correlation to measure each quantity (Hicks and Wesely 1980). The diurnal cycle of sensible heat flux meshes well with expectations based on the available heat energy (i.e., on net radiation), and the friction velocities determined by direct measurement conform to expectations based on the known roughness characteristics of the site. The eddy fluxes of total sulfur demonstrate a diurnal cycle that appears to be as strong as for the meteorological properties, a result that is not surprising when it is remembered that many of the causative factors are common (e.g., vertical turbulent exchange). The quality of the data appears to be quite good--sensible heat fluxes and friction velocities are difficult to measure, and the ability to measure each with the run-to-run smoothness evident in the diagram instills considerable confidence in the sulfur data, measured with the same apparatus. Nevertheless, it would be unwise to place too much confidence in the fine details of the sulfur-flux data. While the strong (downward) fluxes of sulfur evident during the midday periods are probably accurately represented, it is possible that the indicated nighttime fluxes are contaminated by a water-vapor interference or some other factor that is especially significant in the calmer, more humid nighttime situation. Thus, some caution must be associated with interpreting the negative (upward) fluxes of sulfur evident on two periods as evidence of emission or resuspension from the

TABLE C.2-5 Recent Experiments on Trace-Gas Deposition to Natural Surfaces

Worker	Method	Results and Comments
SO_2		
Hill (1971)	$^{35}SO_2$ with stable SO_2 carrier over alfalfa	v_d = 2.3 cm/s in daytime; Implies r_c = 0.4 s/cm
Garland et al. (1973)	$^{35}SO_2$ over pasture	v_d = 1.2 cm s^{-1} in daytime r_c = 0.6 s cm^{-1}
Owers and Powell (1974)	$^{35}SO_2$ over pasture	v_d = 1.3 cm/s, daytime
Shepherd (1974)	SO_2 gradients over grass	v_d= 1.3 cm/s in summer = 0.3 cm/s in autumn
Whelpdale and Shaw (1974)	SO_2 gradients over snow, water, and grass	v_d = 1cm/s in daytime for grass and water snow
Garland (1977)	SO_2 gradients, calcareous soils	v_d = 1.2 cm/s r_c = 0.01 s/cm
Fowler (1978)	SO_2 gradients, over	
	-wheat	v_d = 0.4 cm/s
	-soybean	v_d = 1.3 cm/s

Dannevik et al. (1976)	SO_2 gradients over wheat	$v_d \approx 0.4$ cm/s
Garland and Branson (1977)	$^{35}SO_2$ to pine	$v_d \approx 0.1 - 0.6$ cm/s
Belot (as summarized by Chamberlain 1980)	$^{34}SO_2$ to pine	$v_d < 1$ cm/s
Galbally et al. (1979)	Eddy correlation over pine forest	$v_d \approx 0.2$ cm/s
Dovland and Eliassen (1976)	Accumulation to snow	$v_d \approx 0.1$ cm/s
Barrie and Walmsley (1978)	Accumulation to snow	$v_d \approx 0.2$ cm/s
NO_2		
Wesely et al. (1982)	Eddy correlation - soybeans	$v_d \approx 0.6$ cm/s in daytime $r_c \approx 1.3$ s/cm in daytime; ≈ 15 s/cm at night
O_3		
Galbally and Roy (1980)	Gradients over wheat	$v_d \approx 0.7$ cm/s Implies $r_c \approx 1.4$ s/cm
Wesely et al. (1978, 1982b)	Eddy correlation over a range of natural surfaces	$r_c \approx 0.8$ s/cm in daytime ≈ 1.8 s/cm at night

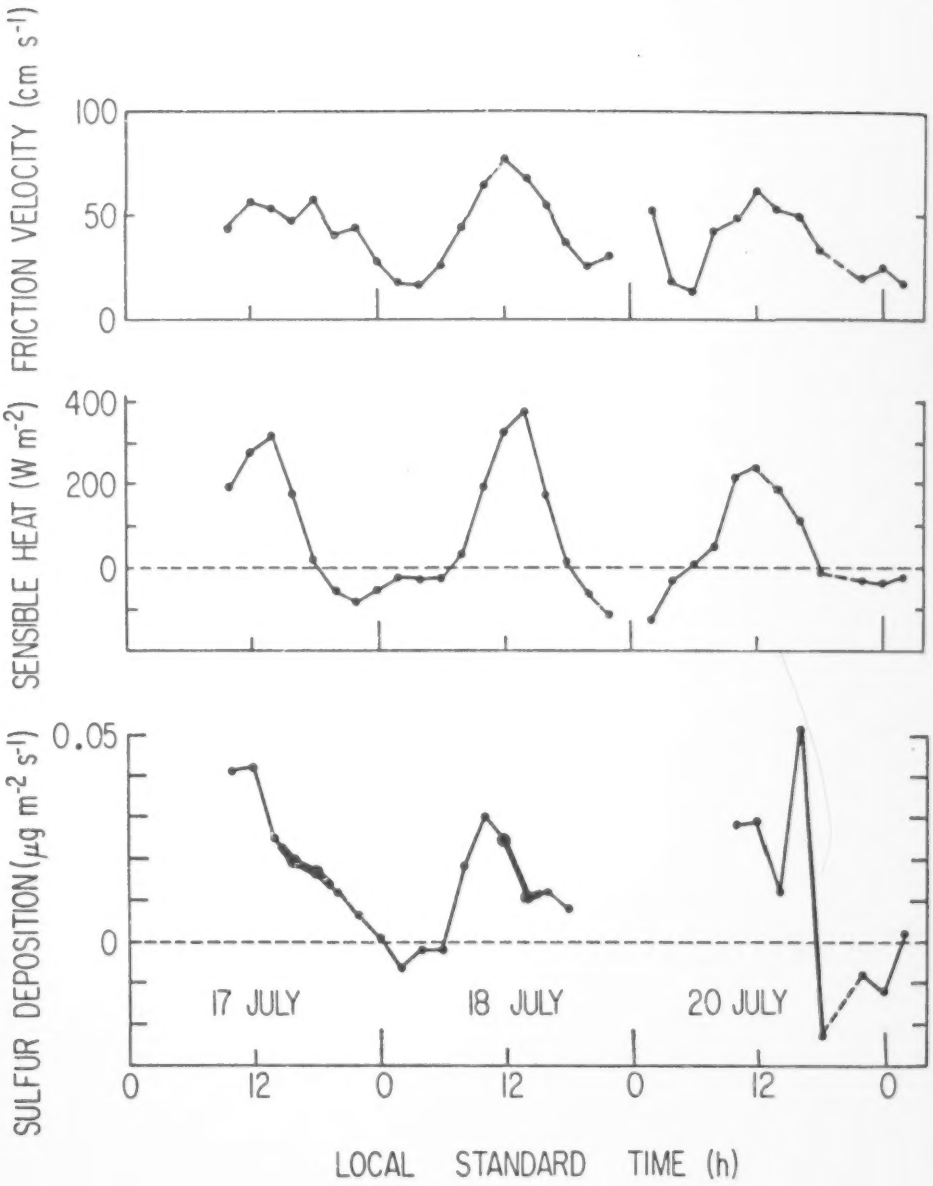

FIGURE C.2-10 Records of sulfur flux, sensible heat flux, and friction velocity through 3 days of an intensive study of dry deposition to a pine plantation (Hicks and Wesely 1980). The darker portions of the sulfur data indicate periods when gaseous sulfur could not be detected. At all times, the data refer to total sulfur, usually made up of gaseous and particulate contributions.

canopy. However, attempts to explain the phenomenon in terms of some interference have so far failed.

Similarly, large diurnal cycles of SO_2 deposition are reported by Fowler (1978), who introduces the further complexity of enhanced SO_2 deposition to wheat covered with dewfall. Using the notation of Figures C.2-5 and C.2-6, Fowler finds typical daytime values to be

$$r_a = 0.25 \text{ s/cm},$$
$$r_b = 0.25 \text{ s/cm},$$
$$r_{st} = 1.0 \text{ s/cm},$$
$$r_{cut} = 2.5 \text{ s/cm}.$$

For deposition to dry soil, Fowler suggests the use of $r_{cs} = 10.0$ s/cm, and $r_{cs} = 0$ when the soil is wet.

It might be noted, in passing, that the aerodynamic resistance r_a influences the deposition of all nonsedimenting pollutants, and thus it is not possible for any trace gas to have a deposition velocity greater than $1/r_a$, i.e., about 4 cm/s in the daytime conditions of Fowler's experiment. Because of stability effects, the maximum possible deposition velocity at night would be considerably lower. Many of the exceedingly large deposition velocities reported in the open literature appear to exceed the limits imposed by our knowledge of the aerodynamic resistance. Thus, several of the results included in the exhaustive tabulation presented by Sehmel (1980b) should be viewed more as indications of experimental error than as determinations of a physical quantity.

Figure C.2-11 addresses the question of the time variation of the aerodynamic resistance, r_a. Values plotted are the maximum deposition velocity permitted by the prevailing aerodynamic resistance, evaluated directly from eddy fluxes of heat and momentum determined during the pine plantation experiment of Figure C.2-10. The reciprocals of the plotted velocities provide evaluations of r_a. It is seen that in daytime, deposition velocities could be as much as 20 cm/s if the surface resistance is zero, implying $r_a = 0.05$ s/cm during midday periods. At night, however, r_a can increase to 10 s/cm on infrequent occasions, but often exceeds 0.5 s/cm. The recommendations of Fowler are probably representative of the long-term average.

The importance of diurnal cycles in pollutant deposition and the close relationship with other meteorological quantities is further illustrated by Figure

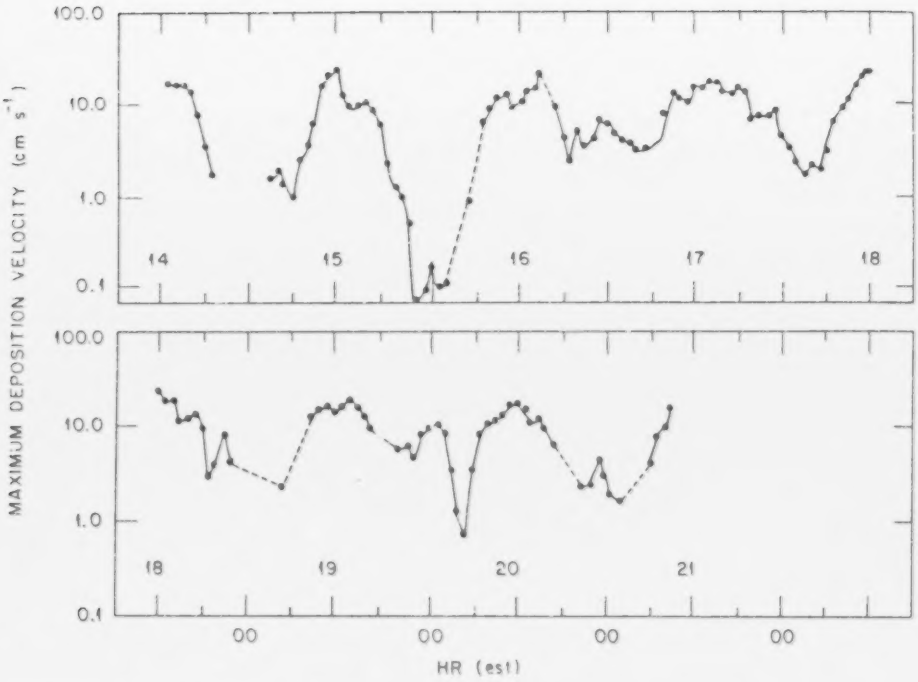

FIGURE C.2-11 Values of the maximum possible deposition velocity of trace gases, determined as the inverse of the aerodynamic resistance r_a for the pine plantation experiment of Hicks and Wesely (1980).

C.2-12, which provides examples of the trend from nighttime, through dawn, and into the afternoon of the residual canopy resistance r_c for ozone and water vapor determined using eddy correlation (Wesely et al. 1978) These data were obtained over corn (Zea mays) in July 1976. The upper sequence shows good matching between r_c for ozone and water vapor, with the former exceeding the latter by a small amount, on the average. As the day progresses, r_c increases gradually, presumably as a consequence of increasing water stress and eventual stomatal closure. The lower data sequence has two features of considerable interest. First, the gradual initial decrease of r_c for O_3 corresponded to a period of evaporation of dewfall (note the relatively low value of r_c for H_2O during the same period), suggesting that the presence of liquid water on the leaf surfaces might inhibit ozone deposition (much as might be expected on the basis of ozone insolubility in H_2O). This would not be the case for SO_2 deposition (Fowler 1978). Second, the peak in both evaluations of r_c at

about 10 a.m. is associated with the passage of clouds, which caused a rapid and strong decrease in incoming radiation and lasted for about an hour. The peak is seen as further evidence for stomatal control, since some stomatal closure would be expected with reduced insolation.

The above discussion of both SO_2 and O_3 deposition confirms the generalization made by Chamberlain (1980) that the deposition of such quantities might be modeled after the case of water-vapor transfer with considerable confidence.

Recently, Wesely et al. (1982b) have reported a field

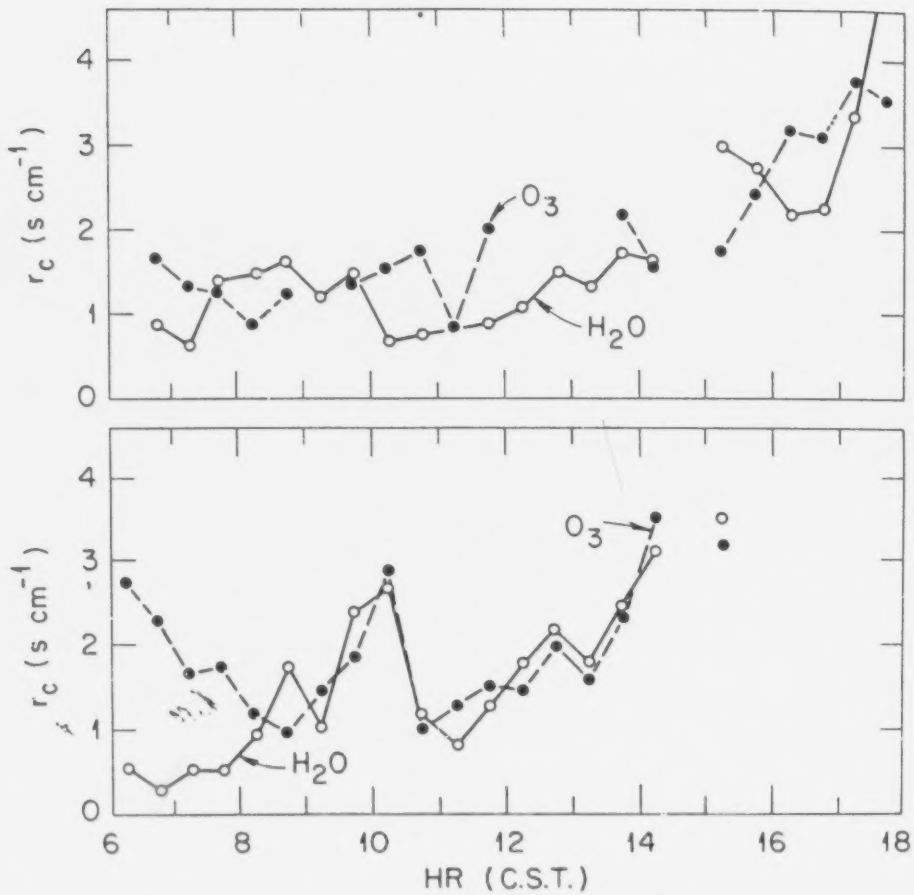

FIGURE C.2-12 Evaluations of the residual "canopy resistance," r_c, to the transfer of ozone and water vapor, based on eddy fluxes measured above mature corn in central Illinois on 29 July 1976 (upper sequence) and 30 July 1976 (lower sequence). Data are from Wesely et al. (1978).

study in which both O_3 and NO_2 fluxes were measured. Bulk canopy resistances to ozone uptake for a soybean canopy exceeded water-vapor values by about 0.5 cm/s during daytime, with r_c for NO_2 still greater by a similar amount.

2.3.2 Particulate Pollutants

No technique for measuring particle fluxes has been developed to the extent necessary to provide universally accepted data. In every case, research and development are continuing at this time. Use of gradient methods, for example, is limited by the inability to resolve concentration differences of the order of 1 percent. Turbulence methods require rapid response, yet sensitive chemical sensors that are not often available. In both cases, practical application is hindered by the need for a site meeting stringent micrometeorological criteria. Nevertheless, several applications of micrometeorological flux-measuring methods have been published. Table C.2-6 provides a list that illustrates the narrow range of available information. The evidence points to a difference between the deposition characteristics of small particles and sulfate; the latter seems to be transferred with deposition velocities somewhat greater than the value of 0.1 cm/s that has been assumed in most assessment studies and greater than the values appropriate for small particles, on the average. At this time, the possibility that sulfate fluxes are promoted by the strong effect of a few large particles cannot be dismissed.

As must be expected, taller canopies are associated with higher values of v_d, on the average. Figure C.2-13 shows how small particle fluxes varied with time of day over a pine plantation in North Carolina during 1977 (Wesely and Hicks 1979). These eddy-correlation results display a run-to-run smoothness that engenders considerable confidence; moreover, they are supported by the finding that simultaneous eddy fluxes of momentum and heat closely satisfied the usual surface roughness and energy balance constraints. There is little doubt that the surface under scrutiny (or at least the air below the sensor) did indeed represent a source rather than a sink for substantial periods (Arnts et al. 1978). A basic question then arises about the significance of the measured deposition rates, since these probably represent a net

TABLE C.2-6 Field Experimental Evaluations of the Deposition Velocity of Submicrometer Diameter Particles

Surface	Size and Method	Results and Comments
Snow		
Dovland and Eliassen (1976)	Lead aerosol, surface sampling	0.16 cm/s in stable stratification, greater values in neutral. All light-wind data
Wesely and Hicks (1979)	0.05-0.1 μm particles eddy correlation	Net fluxes small but upwards; v_d too small to be determined
Open Water		
Sievering et al. (1979)	0.2-1.0 μm particles, gradients	Gradients highly variable. Range of v_d typically 0.2-1.0 cm/s in magnitude. Including reversed gradients in long-term average reduces average v_d to near zero (see Hicks and Williams 1979)
Williams et al. (1978)	0.05-0.1 μm particles, eddy correlation	Preliminary indications only: v_d very small, 95% certainty < 0.05 cm/s
Bare Soil		
Wesely and Hicks (1979)	0.05-0.1 μm particles, eddy correlation	Surface frequently a source v_d very low, on the average but often large for short periods
Grass		
Sehmel et al. (1973b)	Polydispersed rhodamine-B particles with mass median diameter 0.7 μm, deposited to artificial grass exposed outdoors	Average v_d = 0.2 cm/s

TABLE C.2-6 (Continued)

Surface	Size and Method	Results and Comments
Chamberlain (1960)	Radon daughters deposited to natural grass. Work attributed to Megaw and Chadwick	v_d = 0.20 cm/s
Hudson and Squires (1978)	Cloud condensation nuclei fluxes measured by gradient methods over sagebrush and grass. Particle size probably 0.002-0.04 μm	v_d = 0.04 cm/s
Davidson and Friedlander (1978)	~0.03-μm particles, gradients over wild oats	Average v_d = 0.9 cm/s
Wesely et al. (1977)	0.05-0.1 μm particles, eddy correlation	Direction of flux sometimes changes. During deposition periods, v_d = 0.8 cm/s, but much lower on the average
Everett et al. (1979)	Particulate lead and sulfur, gradients	v_d greater for sulfur (~1 cm/s) than for lead from more local sources
Sievering (1982)	0.15-0.3 μm particle gradients over mature rye and wheat	v_d averaged 0.4 ± 0.3 cm/s in light winds, unstable stratification
Hicks et al.(1982)	Sulfate by eddy correlation	v_d as high as 0.7 cm/s in daytime, about 0.2 cm/s as a long-term average
Wesely et al. (1982)	Sulfate by eddy correlation	v_d largest for daytime lush grass (~0.5 cm/s), much less for short dry grass (~0.2 cm/s), strongly stable conditions

Crops		
Droppo (1980)	Particulate trace metals, gradients: senescent maize	v_d varying widely with element, ranging up to about 1 cm/s
Wesely and Hicks (1979)	0.05-0.1 μm particles, eddy correlation: senescent maize	Strong diurnal variation in the direction of the flux. Long-term average $v_d \approx 0.1$ cm s^{-1}
Trees		
Hicks and Wesely (1978, 1980)	Sulfate particles, eddy correlation, Loblolly pine	Strong diurnal variability but less marked than for small particles; average $v_d \approx 0.7$ cm/s
Wesely and Hicks (1979)	0.05-0.1 μm particles, eddy correlation	Very strong diurnal variation with the canopy a net source. During deposition periods, v_d probably greater than 0.6 cm s^{-1}
Lindberg et al. (1979)	Pb, Cd, S, etc. particles, foliar washing	$v_d > 0.1$ cm/s for all quantities, on the average
Wesely et al. (1982)	Sulfate particles, eddy correlation	v_d not significantly different from zero for a winter deciduous forest

result of continuing but varying surface emission and a deposition flux that is also varying with time. The relevance of the answers obtained is then unclear. In particular, it is not obvious how to relate such results to the common situation in which we wish to evaluate the atmospheric deposition rate of some particulate pollutant that is not emitted or resuspended from the surface.

Figure C.2-13 identifies periods of the 1977 pine plantation study during which no gaseous sulfur was detectable. These occasions were used by Hicks and Wesely (1978) to evaluate residual canopy resistances for particulate sulfur that averaged about 1.5 cm/s (with a standard error margin of about ±15 percent) for July 17 and about 1.1 cm/s (±25 percent) for July 18.

Results of two tests of sulfate gradient equipment over arid grassland, reported by Droppo (1980), yield values of 0.10 and 0.27 cm/s for v_d, in very light winds (≈1 m/s). The residual surface resistances evaluated from his data are 7.7 and 3.3 cm/s, respectively. These values are considerably higher than the pine plantation results quoted above but might not be wholly discordant when the nature of the surface present in the gradient studies is taken into account.

Results of an extensive series of eddy-correlation measurements of particulate sulfur fluxes to a variety of vegetated surfaces have been summarized by Wesely et al. (1982a). In daytime conditions, deposition velocities to grass range from about 0.2 to 0.5 cm/s. Values for a deciduous forest in winter (few leaves) are not significantly different from zero. In general, somewhat lower values are appropriate at night. In almost all of the cases summarized by Wesely et al., normalization of surface transfer conductances by u_* appears to reduce the residual variance. Hicks et al. (1982) present supporting data from another study of the same series, also over grassland.

Considerable controversy remains concerning the value of v_d appropriate for formulating the deposition of sulfate aerosol (and presumably all similar particles). Garland (1978) advocates the continued use of values of 0.1 cm/s or less, since experiments conducted over grass in England failed to detect a significant gradient. However, some of the experiments listed in Table C.2-6 indicate quite high deposition velocities for sulfate particles. A possible explanation in terms of a strong contribution by particles much larger than the usual accumulation size mode has been discussed (Garland 1978), and different deposition velocities (0.025 and 0.56 cm/s)

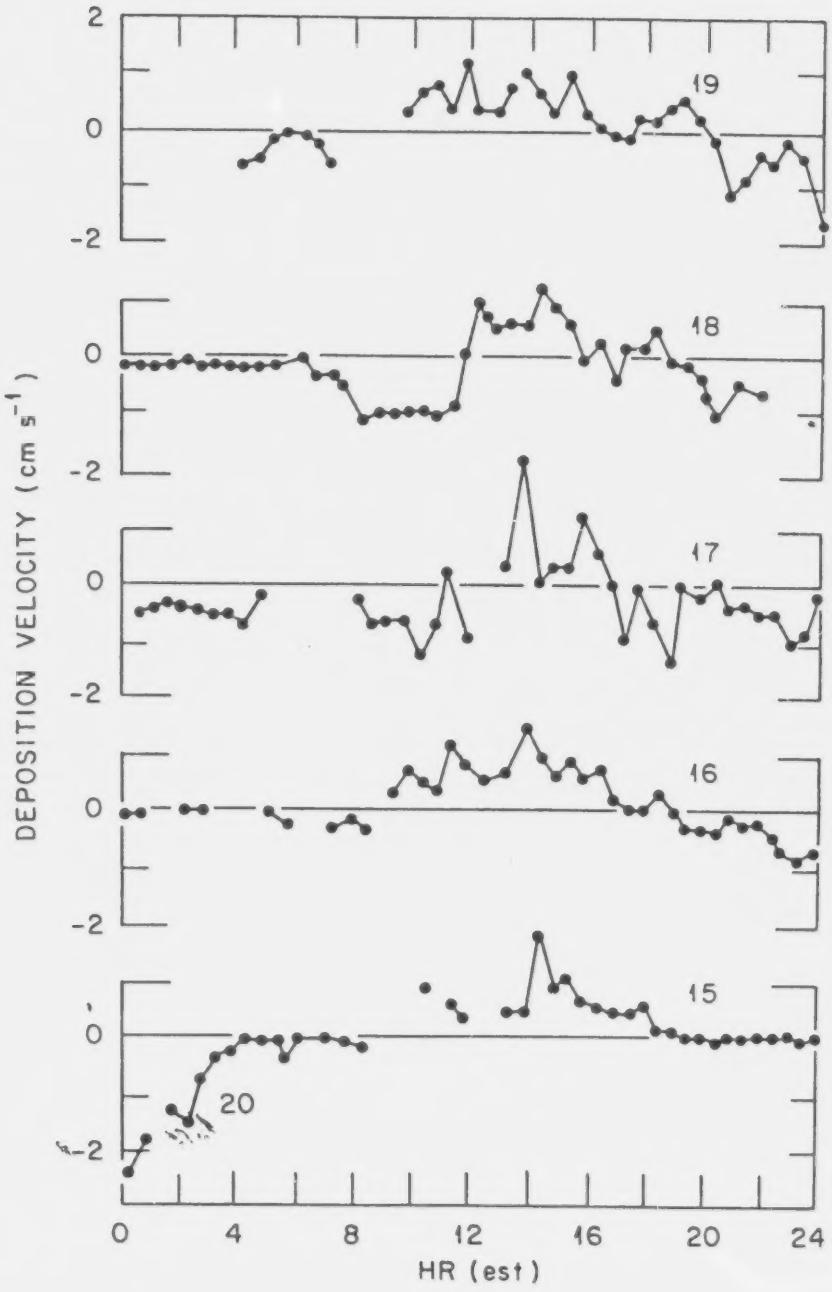

FIGURE C.2-13 Deposition velocity of small particles (~0.1 μm) measured by eddy correlation above a pine plantation in North Carolina in 1977 (Hicks and Wesely 1978, Wesely and Hicks 1979). Note the strong diurnal cycle, with frequent extended periods of emission rather than deposition (as indicated by the negative "deposition velocities").

have been postulated for the submicrometer and larger particles, respectively.

There are great uncertainties about results obtained by deposition plates or other surrogate collection surfaces. All such methods assume that the collection characteristics of some artificial surface are the same as those of the natural surface of interest. Clearly, this assumption will be valid when particles are sufficiently large that gravity is the controlling factor. However, small particles are transferred predominantly by turbulence, with subsequent impaction on the surface of microscale surface roughness elements; these latter factors are not easily reproduced by commonly used artificial collecting devices. The use of collection vessels to monitor the accumulation of particles in them continues to be a widespread practice; however, relating the data obtained to natural circumstances is difficult (q.v. Hicks et al. 1981). In a special category of its own, however, is the method of foliar washing, as used by Lindberg et al. (1979). As applied in careful studies of particle dry deposition at the Walker Branch Watershed in Tennessee, this method of removing and analyzing material deposited on vegetation has succeeded in demonstrating long-term average values of v_d larger than the usually accepted values for several elements.

2.4 MICROMETEOROLOGICAL MODELS OF THE DRY-DEPOSITION PROCESS

2.4.1 Gases

Almost all models of dry deposition of trace gases have as their foundation either the resistance analogy illustrated in Figures C.2-5 and C.2-6 or some equivalent to it. The convenience of this approach is obvious: it permits separate processes to be formulated and combined in a manner that mimics nature, while providing a clear-cut mechanism for determining which processes can be omitted from consideration in specific circumstances. The relevance of the resistance approach to the matter of particle deposition is not so obvious, especially when gravitational settling must be considered.

It is useful to start by identifying the gaseous properties of interest for gases and to identify possible controlling processes.

SO_2: Uptake by plants is largely via stomates during daytime, with about 25 percent apparently via the epidermis of leaves (Fowler 1978). At night, stomatal resistance will increase substantially, but cuticular resistance should be unchanged. When moisture condenses on the depositing surface, associated resistances to transfer should be allowed to decrease to near zero (Fowler 1978, Murphy 1976). To a liquid-water surface, water-vapor appears to provide an acceptable analogy to SO_2 flux.

O_3: Behavior is like SO_2, but with significant cuticular uptake at night ($r_{cut} \sim 2$-2.5 cm/s at night, see r_c quoted by Wesely et al. 1982) and with surface moisture effectively minimizing uptake. Deposition to water surfaces, in general, is very slow.

NO_2: Similar to O_3 in overall deposition characteristics, but with a significant additional resistance (possibly mesophyllic, see Wesely et al. 1982b) of about 0.5 cm/s. Even though NO_2 is insoluble in water in low concentrations, deposition to water surfaces might be quite efficient. Chamber studies (Table C.2-4) indicate similar overall surface resistances for SO_2 and NO_2.

NO: Typical canopy resistances are in the range 5-20 cm/s, as indicated by chamber studies (Table C.2-4) and field experiments (Wesely et al. 1982b). NO appears to be emitted by surfaces at times, possibly as a consequence of NO_2 deposition and of the intimate linkage with ozone concentrations (Galbally and Roy 1980).

HNO_3: No direct information is available; however, on the basis of its high solubility and chemical reactivity, substantial similarity to HF should be expected. Consequently, the use of $r_c = 0$ appears to be a reasonable first approximation.

NH_3: Again, no direct measurements are available but in this case similarity with SO_2 appears likely. Natural surfaces may be emitters of NH_3 because of a number of biological processes occurring in and on soil.

Variations in aerodynamic resistance must be expected to modulate all the behavior patterns summarized above. In many circumstances, deposition rates at night will be nearly zero solely because atmospheric stability is so great that material cannot be transferred through the lower atmosphere. The evaluations given in Figure C.2-12 are especially informative, since even over a pine forest whose surface roughness operates to maximize v_d, an occasion was encountered on one evening out of eight in which atmospheric stability was sufficient to constrain the deposition velocity of all airborne material to less than 0.1 cm/s (with the exception of gravitationally settling particles).

Some models focus on micrometeorological aspects of the pollutant transfer question, others on the biological. Meteorological models tend to follow the lead of agricultural workers. Shreffler (1976) considers profiles of pollutant concentration above and through a canopy, making use of the familiar concepts of zero-plane displacement, leaf area density, and aerodynamic roughness. Interfacial transfer is formulated as recommended by Brutsaert (1975a). The results are shown to agree, in general form, with the experimental data of Chamberlain (1966, for Th-B).

Roth (1975) also uses micrometeorological relations to investigate gaseous dry deposition, adopting the general resistance format and emphasizing the time dependency of the aerodynamic resistance and surface (i.e., canopy) resistance.

Brutsaert (1975b) applies theory to cases of sensible heat and water vapor and extends them to consider a general scalar quantity. He emphasizes that the common micrometeorological assumption that the roughness length is the same for all quantities can lead to considerable error.

O'Dell et al. (1977) consider details of leaf uptake mechanisms, with emphasis on stomatal and mesophyllic resistances. The model addresses questions of plant physiology in detail and is intended to permit comparison of uptake rates of different gaseous species, once air concentrations near leaves are known.

None of these above models considers all the processes that have been discussed above, nor is it likely that any model will have sufficient generality to permit its use for all pollutants in all circumstances. To circumvent many of the difficulties involved, Sheih et al. (1979) prepared a land-use map of North America and coupled this with information regarding surface conditions in order to

derive a spatial distribution of surface resistances appropriate for formulating the deposition of sulfur dioxide. By coupling these values with aerodynamic resistances characteristic of different stability conditions and for different seasons, they produced a map of SO_2 deposition velocities suitable for use in numerical models and for interpreting concentration data. This approach has not been extended to other gaseous pollutants.

2.4.2 Particles

Modeling of particle deposition is complicated by three major factors: (1) gravitational settling, which causes particles to fall through the atmospheric turbulence that provides the conceptual basis for conventional micrometeorological models (Yudinge 1959); (2) particle inertia, which permits particles to be projected through the near-surface laminar layer by turbulence but also prohibits them from responding to the high-frequency turbulent motions that transport material near receptor surfaces; and (3) uncertainty regarding the processes that control particle capture. These three factors are interrelated in such a manner that clear-cut differentiation of their separate consequences is not possible.

The problem has attracted the attention of many theoreticians, and many numerical models have been developed. Each model represents a selected combination of processes, chosen for consideration on the basis of the modeler's understanding of the problem. Without adequate consideration of all the mechanisms involved, none of these models can be considered as a simulator of natural behavior. This is not to question the worth of such models, but rather to emphasize that each should be applied with caution and only to those situations commensurate with its own assumptions.

The many numerical models can be classified in several different ways. Some extend chemical engineering results to surface geometries that are intended to represent plant communities. Others extend agrometeorological air-canopy interaction models by including critical aspects of aerosol physics. Both approaches have benefits, and the final solution will probably include aspects of each.

An excellent review of model assumptions has been given by Davidson and Friedlander (1978). They trace the evolution of models from the 1957 work of Friedlander and Johnstone, which concentrated on the mechanism of inertial

impaction and assumed that particles shared the eddy diffusivity of momentum, to the canopy filtration models of Slinn (1974) and Hidy and Heisler (1978). Early work concerned deposition to flat surfaces and made various assumptions about the surface collection process. Friedlander and Johnstone (1957) permitted particles to be carried by turbulence to within one free-flight distance of the surface, upon which they were assumed to be impacted by inertial penetration of the quasi-laminar "viscous" sublayer. Beal (1970) introduced viscous effects to limit the transfer of small particles, while retaining inertial impaction of larger particles. Sehmel (1970) assumed that all particles that contact the surface will be captured and used empirical evidence obtained in his wind-tunnel studies to determine the overall resistance to transfer, assumed to apply at a distance of one particle radius from the surface. Sehmel's work has been updated recently to provide an estimate of deposition velocities to canopies of a range of geometries in different meteorological conditions (see Sehmel, 1980a).

The above models are based largely on observations and theory regarding the deposition of particles to smooth surfaces, usually of pipes. More micrometeorologically oriented models have been presented by workers such as Chamberlain (1967), who extended the familiar meteorological concepts of roughness length and zero plane displacement to the case of particle fluxes. Much of this work was considered as an extension of models developed for the case of gaseous deposition to vegetation, which in turn were based on an extensive background of agricultural and forest meteorology, especially concerning evapotranspiration. A recent development of this genre is the canopy model of Lewellen and Sheng (1980), which utilizes recent techniques in turbulence modeling to reproduce the main features of subcanopy flow and combines these with particle-deposition formulations like those represented in Figure C.2-1. Lewellen and Sheng emphasize their model's omission of several potentially critical mechanisms, especially electrical migration, coagulation, evolution of particle size distributions, diffusiophoresis, and thermophoresis. To this list we can add a number of other factors about which little is known at this time such as subcanopy chemical reactions, interactions with emissions, and the effect of microscale roughness elements.

Although outwardly simpler than the case of particle deposition to a canopy, that to a water surface has given rise to a similar variety of models. Once again, however, different models focus on different mechanisms. That of Sehmel and Sutter (1974) is based on their wind-tunnel observations and lacks a component that can be identified with wave effects. Slinn and Slinn (1980) invoke the rapid growth of hygroscopic aerosol particles in very humid air to propose rather rapid deposition to open water; deposition velocities of the order of 0.5 cm/s appear possible in this case. On the other hand, Hicks and Williams (1979) propose negligible fluxes unless the surface quasi-laminar layer is interrupted by breaking waves. At present, none of these models has strong experimental evidence to support it. However, experimental and theoretical studies are proceeding, and a resolution of the matter can certainly be expected.

2.5 CONCLUSIONS TO SECTION 2

All the many processes that combine to permit airborne materials to be deposited at the surface have aspects that are strongly surface dependent. While broad generalities can be made about the velocities of deposition of specific chemical species in particular circumstances, there will be wide temporal and spatial variabilities of most of the controlling properties. The detailed nature of the vegetation covering the surface is often a critical consideration. If depositional inputs to some special sensitive area need to be estimated, then this can only be accomplished if characteristics specific to the vegetation cover of the area in question are taken into account in an adequate manner.

Recent field studies investigating the fluxes of small particles have confirmed wind-tunnel results that point to a surface limitation. Studies of the rate of deposition of particles to the internal walls of pipes and investigations of fluxes to surfaces more characteristic of nature, exposed in wind tunnels, tend to confirm theoretical expectations that surface uptake is controlled by the ability of particles to penetrate a quasi-laminar layer adjacent to the surface in question. The mechanisms that limit the rate of transfer of particles involve their finite mass. Particles fail to respond to the high-frequency turbulent fluctuations that cause transfer to take place in the immediate vicinity of a surface. However, the momentum of particles also causes

an inertial deposition phenomenon that serves to enhance the rate of deposition of particles in the 10- to 20-μm size range.

The general features of particle deposition to smooth surfaces are fairly well understood. Studies conducted so far support the theoretical expectation that particles smaller than about 0.1 μm in diameter will be deposited at a rate that is largely determined by Brownian diffusivity. In this instance, the limiting factor is the transfer by Brownian motion across the quasi-laminar layer referred to above. On the other hand, particles larger than about 20 μm in diameter are effectively transferred via gravitational settling, at rates determined by the familiar Stokes-Cunningham formulation. Particles in the intermediate size ranges are transferred very slowly. The minimum value of the "well" of the deposition velocity versus particle size curve is approximately 0.001 cm/s.

However, natural surfaces are rarely aerodynamically smooth. Wind-tunnel studies have shown that the "well" in the deposition velocity curve is filled in as the surface becomes rougher. Although studies have been conducted, in wind tunnels, of deposition fluxes to surfaces such as gravel, grass, and foliage, the situation involving natural vegetation such as corn, or even pasture, remains uncertain. It is well known that many plant species have foliage with exceedingly complicated microscale surface roughness features. In particular, leaf hairs increase the rate of particle deposition; studies of other factors, such as electrical charges associated with foliage and stickiness of the surface, indicate that a natural canopy might be considerably different from the simplified surfaces suitable for investigation in laboratory and wind-tunnel investigations.

Caution should be exercised in extending laboratory studies using artificially produced aerosol particles to the situation of the deposition of acidic quantities. Special concern is associated with the hygroscopic nature of many acidic species. Their growth as they enter a region of high humidity and their liquid nature when they strike the surface are both potentially important factors that might work to increase otherwise small deposition velocities. Moreover, there is evidence that acidic particles, especially sulfates, might be carried by larger particles; the rates of deposition of such complicated particle structures are essentially unknown. However, the shape of particles can have a considerable

influence on their gravitational settling speed and probably on their impaction characteristics as well.

It is not clear to what extent special considerations appropriate for acidic species, such as those mentioned above, contribute to the finding of unexpectedly high deposition velocities for atmospheric sulfate particles, as reported in some recent North American studies. European work has been fairly uniform in producing deposition velocities close to 0.1 cm/s, while North American experience has generated larger values.

It is informative to consider the flux of any airborne quantity to the surface underneath in terms of an electrical analog, the so-called resistance model developed initially in studies of agrometeorology. In this model, the flux of the atmospheric property in question is identified with the flow of current in an electrical circuit; individual resistances can then be associated with readily identifiable atmospheric and surface properties. While the electrical analogy has obvious shortcomings, it permits an easy visualization of many contributing processes and enables a comparison of their relative importance. Micrometeorological studies of the fluxes of atmospheric heat and momentum show that the aerodynamic resistance to transfer (i.e., the resistance to transfer between some convenient level in the air and a level immediately above the quasi-laminar layer) ranges from between 0.1 s/cm in strongly unstable, daytime conditions, to more than 10 s/cm in many nocturnal cases. There are several resistance paths that permit gaseous pollutants to be transferred into the interior of leaves. An obvious pathway is directly through the epidermis of leaves, involving a cuticular resistance. An alterhative route, known to be of significantly greater importance in many cases, is via the pores of leaves, involving a stomatal resistance that controls transfer to within stomatal cavities and a subsequent mesophyllic resistance that parameterizes transfer from substomatal cavities to leaf tissue. Comparison between resistances to transfer for water vapor, ozone, sulfur dioxide, and gases that are similarly soluble and/or chemically reactive shows that in general such quantities are transferred via the stomatal route, whenever stomates are open. Otherwise, cuticular resistance appears to play a significant role. Cuticular uptake of ozone and of quantities like NO and NO_2 appears to be quite significant, whereas for SO_2 this pathway appears to be less important. When leaves are wet, such as after heavy dewfall, uptake of sulfur dioxide is exceedingly efficient

until the pH of the surface water becomes sufficiently acidic to impose a chemical limit on the rate of absorption of gaseous SO_2. However, the insolubility of ozone causes dewfall to inhibit ozone dry deposition.

The same conceptual model can be applied to the case of particle transfer with considerable utility. While the roles of factors such as stomatal opening become less clear when particles are being considered, the concept of a residual surface resistance to particle uptake appears to be rather useful. Studies of the transfer of sulfate particles to a pine forest have shown that this residual surface resistance is of the order of 1 to 2 s/cm. It appears probable that substantially larger values for residual surface resistance will be appropriate for nonvegetated sufaces, especially to snow, for which the values are more likely to be approximately 15 s/cm. At this time, an exceedingly limited quantity of field information is available; however, it appears that in North American conditions the surface resistance to uptake of sulfate particles will be in the range 1.5 to 15 s/cm.

While sulfate particles have received most of the recent emphasis, the general question of acid deposition requires that equal attention be paid to nitrate and ammonium particles. There is little information regarding the deposition velocities of these particles. However, there is no strong indication that they are different from the case of sulfate.

Regarding trace-gas uptake, sulfur dioxide has received the majority of recent attention. Chamber studies and some recent field work indicate that highly reactive materials such as hydrogen fluoride (and presumably iodine vapor, nitric acid vapor, etc.) are readily taken up by a vegetative surface, whereas a second set of pollutants including SO_2, NO_2, and O_3 seem to be easily transferred via stomates, and a third category of relatively unreactive trace gases are poorly taken up.

Transfer to water surfaces presents special problems, especially when the surface concerned is snow. As mentioned above, surface resistances to particle uptake by snow appear to be of the order 15 s/cm. Soluble gases will be readily absorbed by all water surfaces, and so equivalence to transfer of water vapor might be expected. An important exception occurs in the case of SO_2, in which case absorbed SO_2 can increase the acidity of the surface moisture layer to the extent that further SO_2 transfer is cut off. Trace-gas transfer to liquid-water surfaces is influenced by the Henry's law constant.

Wind-tunnel studies of particle transfer to water surfaces all show exceedingly small deposition velocities for particles in the 0.1- to 1-μm size range. Several workers have suggested mechanisms by which larger deposition velocities might exist in natural circumstances; for example, the growth of hygroscopic particles in highly humid, near-surface air can cause accelerated deposition of such particles, and breaking waves might provide a route that bypasses the otherwise-limiting quasi-laminar layer in contact with a water surface. Once again field observations are lacking.

While larger deposition velocities of soluble trace gases to open water surfaces appear quite likely, water bodies are frequently sufficiently small that air-surface thermal equilibrium cannot be achieved. Air blowing from warm land across a small cool lake, for example, will not rapidly equilibrate with the smooth, cooler surface. Flow will then be stable and largely laminar, with the consequence that very small deposition velocities will apply for all atmospheric quantities. In many circumstances, especially in daytime summer occasions, deposition velocities are likely to be so small as to be disregarded for all practical purposes. On the other hand, during winter when the land surface is frequently cooler than the water, the resulting corrective activity over small water bodies will cause the air to come into fairly rapid equilibrium with the water, and rather high deposition velocities, in agreement with the open water surface expectations, will probably be attained.

An associated special case concerns the effect of dewfall, which can accelerate the net transfer of trace gases and particles in some circumstances. The velocities of deposition involved are small, however they permit an accumulation of material at the surface in conditions in which the atmospheric considerations are likely to predict minimal rates of exchange (i.e., limited by stability to an extreme extent). When surface fog exists, the highly humid conditions will permit airborne hygroscopic particles to nucleate and grow rapidly. This process provides a mechanism for cleansing the lower layers of the atmosphere of most acidic airborne particles. The small fog droplets that are formed around the hygroscopic acidic nuclei are transferred by the classical process of fog interception to foliage and other surface roughness elements.

Conclusions of recent workshops (e.g., Hicks et al. 1981) have indicated that it is not possible to measure

the dry deposition of acidic atmospheric materials using exposed collection vessels, since they fail to collect trace gases and small particles in a manner that can be related in a direct fashion to natural circumstances. However, surrogate surface methods appear to be useful in indicating space and time variations of deposition in some cases. It is possible to measure the flux of some airborne quantities by micrometeorological means, without interfering with the natural processes involved. These studies, and laboratory and wind-tunnel investigations, provide evidence that the controlling properties in the deposition of many gaseous pollutants are associated with surface structure, rather than with atmospheric properties. The exception to this generalization is the nocturnal case, in which atmospheric stability may often be sufficient to impose a severe restriction on the rate of delivery of all airborne quantities to the surface below.

The conclusions presented above can be summarized as follows:

- Dry deposition of small aerosol particles and trace gases is a consequence of many atmospheric, surface, and pollutant-related processes, any one of which may dominate under some set of conditions. The complexity of each individual process makes it unlikely that a comprehensive simulation will be developed in the near future.
- The convenient simplicity afforded by the concept of a deposition velocity (or its inverse, the total resistance to transfer) makes it possible to incorporate dry-deposition processes in models in a manner that is adequate for modeling and assessment purposes. The simplicity of the deposition velocity approach imposes obvious limitations on its applications. For example, the use of average deposition velocities is inappropriate when it is desired to look at time- or space-resolved details of deposition fluxes.
- Sufficient is known about the processes that control the deposition of trace gases that in many instances deposition velocities can be considered to be known functions of properties such as wind speed, atmospheric stability, surface roughness, and biological factors such as stomatal aperture. Important exceptions are for the case of insoluble (or poorly soluble) gases and for deposition to nonsimple surfaces such as forests in rough terrain.

- The deposition of particles larger than about 20-μm diameter is controlled by gravity and can be evaluated using the Stokes-Cunningham relationship. These large particles might contribute to the deposition of acidic and acidifying substances.
- The deposition of small particles remains an issue of considerable disagreement. On the whole, model predictions agree with the results of laboratory and wind-tunnel studies, at least for test surfaces that are usually smoother than pasture, but field experiments provide data that indicate greater deposition velocities. The reasons for the apparent disagreement are not yet clear.
- Over water surfaces, there are almost no field data on the deposition of small particles. Different models have been put forward, predicting a wide range of deposition velocities. At this time, there is little evidence that would permit us to choose between them. The situation for trace gases like sulfur dioxide and ammonia is much better. On the whole, models agree with the available field data, although there is disagreement between the models on how factors such as molecular diffusivity should be handled.
- Dry deposition to the surfaces of materials used in the construction of buildings and monuments, for example, can be measured in many instances by taking sequential samples of the surface over extended periods.
- Particulate material at the surface can creep, bounce, and eventually resuspend under the influence of wind gusts. The large particles entrained in this way can cause a local modification of the acid deposition phenomenon that is associated with accumulation-size aerosol particles and trace gases of more distant origin.
- For both case-study measurement purposes and for long-term monitoring, accurate measurements of pollutant air concentrations are necessary. For monitoring purposes, measurement of airborne pollutant concentrations in a manner carefully designed to permit evaluation of dry-deposition rates by applying time-varying deposition velocities specific to the pollutant and site in question appears to be the most attractive option.
- Micrometeorological methods for measuring dry-deposition fluxes have been developed from the techniques conventionally used to determine fluxes of sensible heat, moisture, and momentum. These methods are technologically demanding, and their use in routine monitoring applications is not yet possible.

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3. PRECIPITATION SCAVENGING PROCESSES

3.1 STEPS IN THE SCAVENGING SEQUENCE

3.1.1 Introduction

Precipitation Scavenging is defined generally as the composite process by which airborne pollutant gases and particles attach to precipitation elements and thus deposit to the Earth's surface.* This process typically contains many parallel and consecutive steps, and as an introduction to this section it is appropriate to provide a brief overview of these intermeshing pathways. In a general sense there are four major events in which a natural or pollutant molecule† may participate, prior to its wet removal from the atmosphere; depicted pictorially in Figure C.3-1, these are as follows:

1-2. The pollutant and the condensed atmospheric water (cloud, rain, snow, . . .) must intermix within the same airspace.

2-3. The pollutant must attach to the condensed-water elements.

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*One should note that this definition pertains to removal from the gaseous medium of the atmosphere combined with deposition to the ground. An alternative definition, employed often throughout the open literature, pertains to the simple attachment of airborne pollutants to liquid water elements, without regard to whether the material is subsequently conveyed to the Earth's surface. Which of these definitions is used is unimportant so long as the precise definition is understood. The definition of "scavenging" adopted here will be utilized consistently throughout this text. When specific reference to the alternative situation is made, the terms "attachment" and "capture" will be employed essentially interchangeably.

†Initial portions of this section will treat precipitation scavenging in a general sense, with limited reference to specific types of atmospheric material. The reader should continue to note, however, that the "natural or pollutant molecules" of primary concern in the present context are species associated with acid-base formation, such as SO_2, HNO_3, NH_3, sulfate, chloride, and metallic cations.

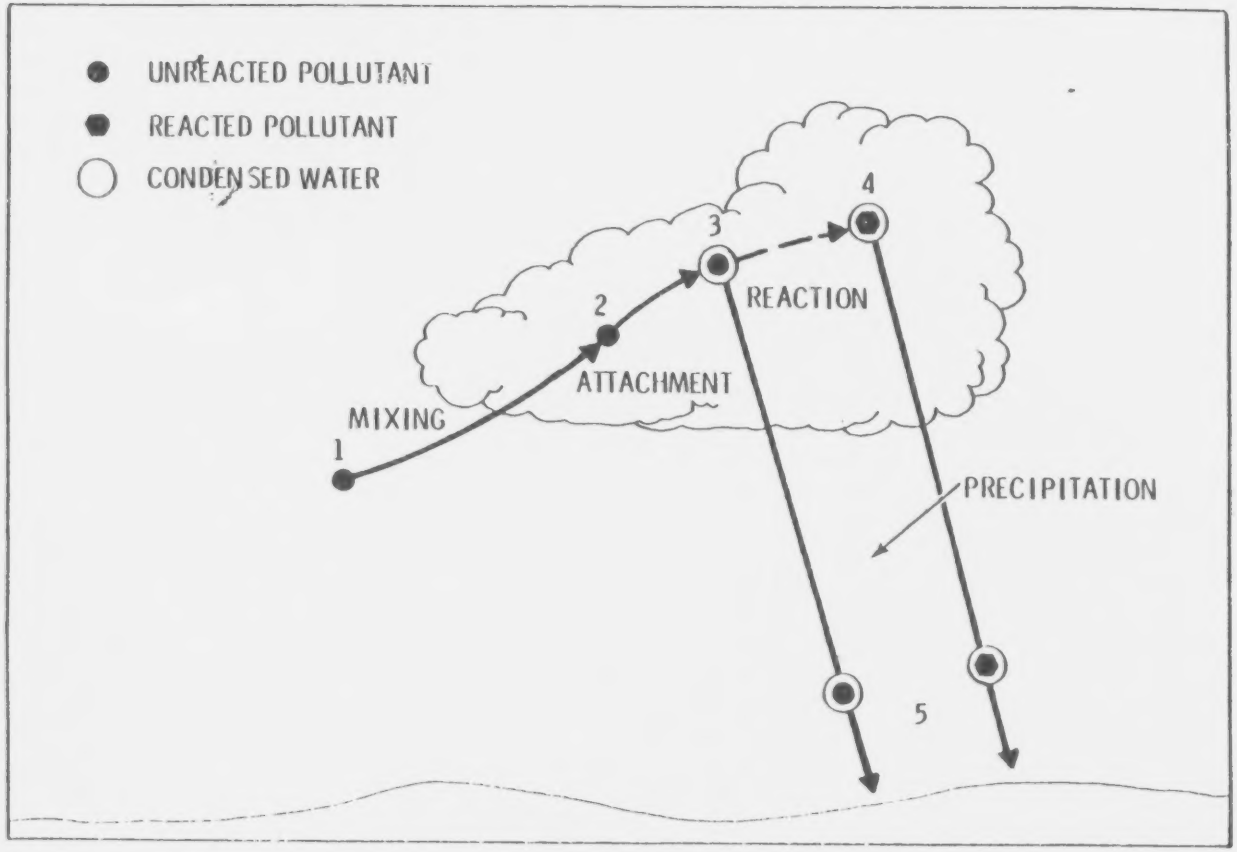

FIGURE C.3-1 Steps in the scavenging sequence: pictorial representation.

3-4. The pollutant may react physically and/or chemically within the aqueous phase.

3-5 or (4-5). The pollutant-laden water elements must be delivered to the Earth's surface via the precipitation process.

The interaction diagram of Figure C.3-2 gives a somewhat more detailed portrayal of these four major events. Here the individual steps are represented as transitions of the pollutant between various states in the atmosphere, and one can note that a multitude of reverse processes is also possible; thus a particular pollutant molecule may experience numerous cycles through this complex of pathways prior to deposition. Indeed, Figure C.3-2 indicates that this cycling process may continue even after "ultimate" deposition. By pollutant off-gassing and other resuspension processes the deposited material can be re-emitted to the atmosphere, with the possibility of participating in yet another series of cycles throughout the scavenging sequence.

Another important feature of Figure C.3-2 is its indication that, while physicochemical reaction within the aqueous phase is potentially an important step in the scavenging process, it is not essential. This contrasts to the remaining forward steps that must take place if scavenging is to occur. Despite its nonessential nature, this step is often of utmost importance in influencing scavenging rates, owing to its role in modifying reverse processes in the sequence. An example of this effect is the devolatilization of dissolved sulfur dioxide via wet oxidation to sulfate. This effectively eliminates gaseous desorption from the condensed water and thus has a strong tendency to enhance the overall scavenging rate as a result.

From Figure C.3-2 one can note also that precipitation scavenging of pollutant materials from the atmosphere is intimately linked with the precipitation scavenging of water. If one were to replace the word "pollutant" with "water vapor" in each of the steps, Figure C.3-2 (with the exception of box 4) would provide a general description of the natural precipitation process. In view of this intimate relationship, it is not surprising that pollutant wet-removal behavior tends to mimic that of precipitation. Pollutant-scavenging efficiencies of storms, for example, are often similar to water-extraction efficiencies. This relationship is useful in the practical estimation of scavenging rates and will

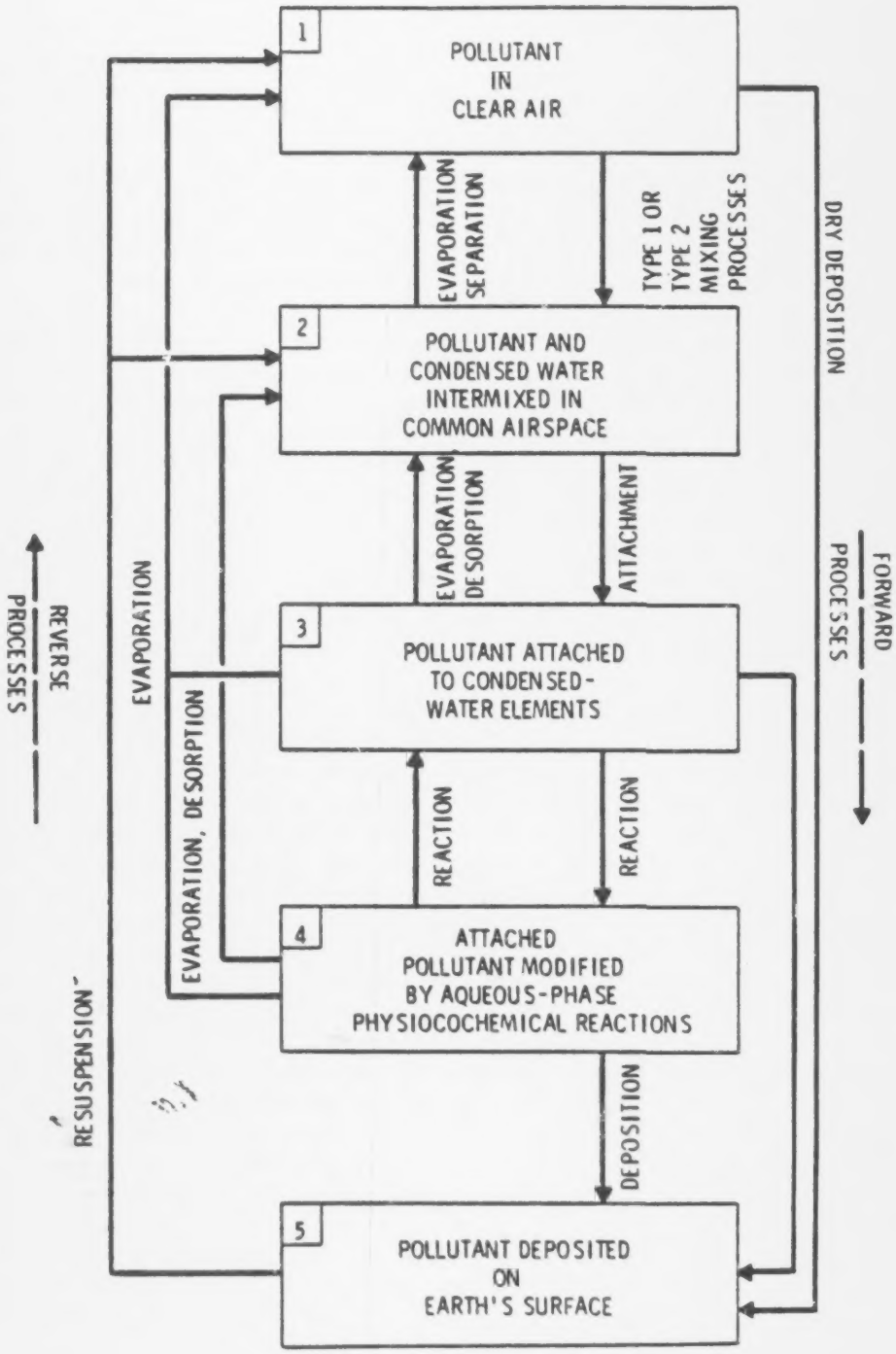

FIGURE C.3-2 Scavenging sequence: interaction diagram.

reappear continually in the ensuing discussion of wet-removal behavior.

Figure C.3-2 is interesting also because of its indication that, if some particular step in the diagram occurs particularly slowly compared with the others, then this step will dominate behavior of the overall process. This is similar to the "rate-controlling step" concept in chemical kinetics and has been applied rather extensively in practical scavenging calculations (Slinn 1974a). Finally, it is important to note that Figure C.3-2 presents a framework for development and evaluation of mathematical models of scavenging behavior. Successful scavenging models must emulate these steps effectively and tend to reflect the structure of Figure C.3-2 as a result. This point will re-emerge later when scavenging models are examined specifically. The remainder of the present subsection will address qualitative aspects of the scavenging sequence, in the order of their forward progress to ultimate deposition.

3.1.2 Intermixing of Pollutant and Condensed Water (Step 1-2)

On first consideration one often is inclined to dismiss pollutant/condensed-water intermixing as an unimportant, or at least trivial, step in the overall scavenging sequence. It is neither. In a statistical sense it usually is neither cloudy nor precipitating in the immediate locality of a freshly released pollutant molecule; and typically this molecule must exist in the clear atmosphere for several hours, or even days, before it encounters condensed water with which it may co-mingle. This in itself establishes step 1-2 as a potentially important rate-influencing event. Moreover, this extended dry period typically presents the pollutant with significant opportunities to react and/or deposit via dry processes; thus the chemical makeup of precipitation is influenced profoundly by this preceding chain of events.

Significant insights into the behavior of step 1-2 can be gained via past analyses of storm formation (e.g., Godske et al. 1957) and the atmospheric water cycle (Newell et al. 1972). Several statistical analyses of precipitation occurrence (Baker et al. 1969; Junge, 1974; Rodhe and Grandell 1972, 1981; Slinn, 1973b) have been applied as general interpretive descriptors of this step. These will not be examined in detail here; rather,

we shall concentrate on the mechanisms by which step 1-2 can occur, from a more pictorial viewpoint.

Two types of mixing processes exist in which pollutant and condensed water can come to occupy common airspace; these are as follows:

1. Relative movement of the initially unmixed pollutant and condensed water, in a manner such that they merge into a common general volume;

and

2. In situ phase change of water vapor, thus producing condensed water in the immediate vicinity of pollutant molecules.

The relative importance of Type-1 and Type-2 mixing processes will depend to some extent on the pollutant. If a particular pollutant is easily scavengable, and if precipitation is occurring at the pollutant's release location, then Type-1 processes are likely to contribute significantly. If these two conditions are not met, the pollutant will usually mix intimately with makeup water vapor for some future cloud, and Type-2 processes will predominate. Based on in-cloud versus below-cloud scavenging estimates (Slinn 1983) it is not unreasonable to estimate that, as a global average, roughly 90 percent of all precipitation scavenging occurs as the consequence of a Type-2 process.

As indicated in Figure C.3-2, reverse processes exist that can serve to reseparate pollutant and condensed water. Evaporation, for example, can reinject pollutant from cloudy to clear air, and relative motion such as precipitation "fall-through" can remove hydrometeors from contact with elevated plumes. Cloud formation-re-evaporation cycles are particularly significant in this respect. Junge (1964), for example, estimates that a single cloud condensation nucleus is likely to experience of the order of ten or more evaporation-condensation cycles before it is ultimately delivered to the Earth's surface with precipitation. The rate-influencing effect of such cycling on precipitation scavenging is obvious. Additional types of cycles will be described below, in conjunction with succeeding steps of the scavenging sequence.

3.1.3 Attachment of Pollutant to Condensed-Water Elements (Step 2-3)

The microphysics of the pollutant-attachment process has been the subject of extensive research, and numerous reviews of this area have been prepared (e.g., Davies 1966, Dingle and Lee 1973, Hales 1983, Junge 1963, Pruppacher and Klett 1978, Slinn 1983, Slinn and Hales 1983). In the context of Figure C.3-1, this process is complicated somewhat in the sense that, depending on the particular attachment mechanism, Step 2-3 may occur either simultaneously or consecutively with Step 1-2.

Simultaneous comixing and attachment occur in the case of cloud-particle nucleation. This is a phase-transformation (Type-2) process wherein water molecules, thermodynamically inclined to condense from the vapor phase, migrate to some suitable surface for this purpose. Pollutant aerosol particles provide such surfaces within the air parcel, and the consequence is a cloud of droplets (or ice crystals)* containing attached pollutant material.

Different types of aerosol particles possess different capabilities to nucleate cloud elements and grow by the condensation process. As a consequence there is typically a competition for water molecules among the aerosol and associated cloud particles; some will capture water with high efficiency and grow substantially in size. Others will acquire only small amounts of water, and still others will remain essentially as "dry" elements. In addition, some particles may nucleate ice crystals, while others will be active only for the formation of liquid water. The nucleating capability of a particular aerosol particle is determined by its size, its morphological characteristics, and its chemical composition. Various

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*At this point it is important to note that aerosols can participate in several types of phase transitions in cloud systems. These include vapor-liquid, vapor-solid, and liquid-solid transitions, in addition to a subset of interactions between numerous solid phases. Particles active as ice-formation nuclei are generally much less abundant than those active as droplet (or "cloud-condensation") nuclei. As will be demonstrated later, the relative abundance of ice nuclei can have a profound effect on precipitation-formation processes and related scavenging phenomena.

aspects of this subject are discussed at length in standard cloud-physics textbooks (e.g., Mason 1971, Pruppacher and Klett 1978) and in the periodical literature (e.g., Fitzgerald 1974).

An additional important aspect of the cloud-droplet nucleation and growth process is the fact that once initiated, cloud-droplet growth does not proceed instantaneously to some sort of thermodynamic equilibrium. Because of diffusional constraints on delivering water molecules from the surrounding atmosphere, the growth in droplet diameter slows appreciably as droplet size increases (cf. Slinn 1983). Superimposition of this lag on the continually fluctuating environment of a typical cloud results in a dynamic and complex physical system.

Finally, the competitive nature of the cloud-nucleation process results in significant impacts by the pollutant on the basic character of the cloud itself. If the local aerosol were populated solely by a relatively small number of large, hygroscopic particles, for example, one would expect any corresponding cloud to be composed chiefly of low populations of large droplets. If on the other hand the local aerosol were composed of large numbers of small, nonhygroscopic particles, the corresponding cloud should contain larger numbers of smaller droplets.

This is precisely what is observed in practice. Unpolluted marine atmospheres, for example, contain large sea-salt particles as a primary component of their aerosol burden. Warm marine clouds are noted for their wide drop spectra containing large drop sizes and their corresponding capability to form precipitation easily. Continental clouds, on the other hand, are typically composed of larger populations of smaller droplets. Figure C.3-3, which was prepared on the basis of results published by Squires and Twomey (1960), provides a good example of this point. Here measured convective-cloud droplet spectra are compared for two different cloud systems. The continental air-mass cloud exhibits a distinct tendency toward smaller drop sizes and larger populations, as compared with its maritime counterpart. It is interesting also in this context to note the estimates of Junge (1963) with regard to relative amounts of aerosol participating in the nucleation process. Junge suggests that while 50 to 80 percent of the mass of continental aerosols can be expected to participate as cloud nuclei, as much as 90 to 100 percent of maritime aerosols can become actively involved.

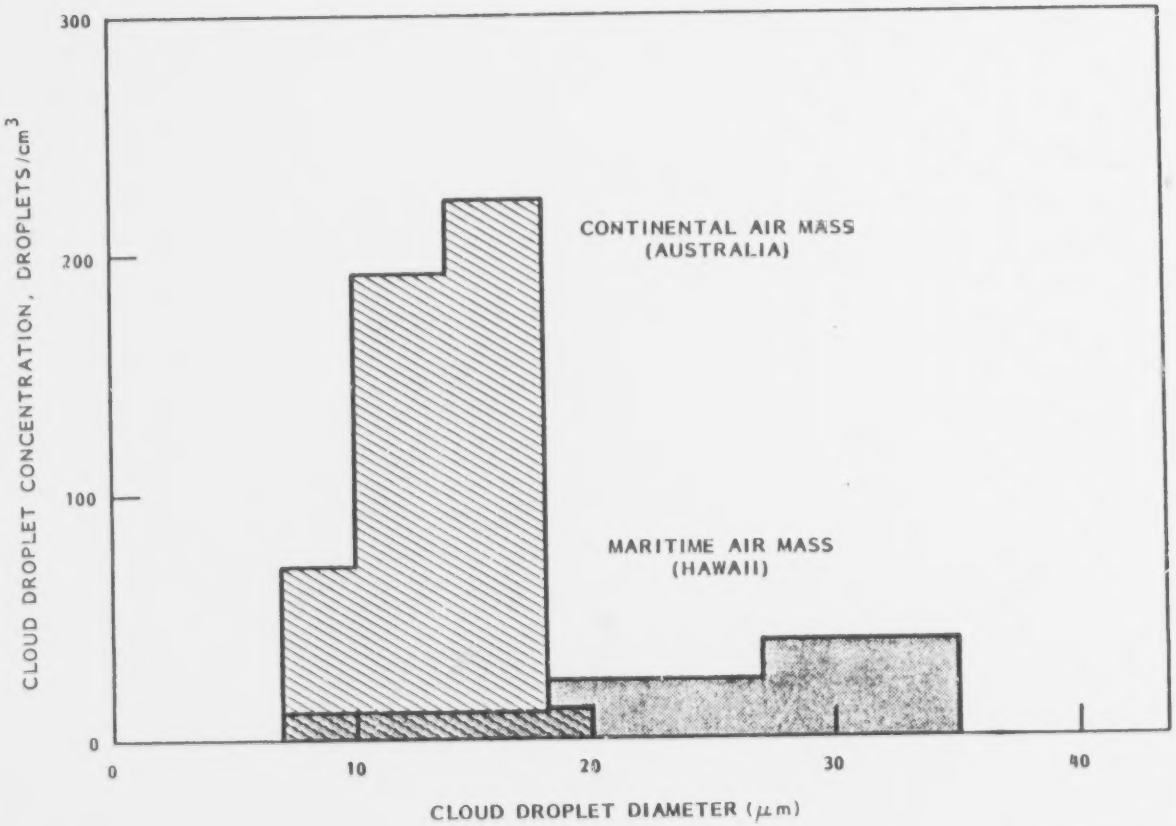

FIGURE C.3-3 Cloud droplet spectra in convective clouds formed in maritime and continental air masses. Adapted from Squires and Twomy (1960).

As a concluding note in the context of nucleating capability and water competition it should be pointed out that acid-forming particles, by their very nature, are chemically competitive for water vapor and thus tend to participate actively as cloud-condensation nuclei. This attribute tends to enhance their propensity to become scavenged early in storm systems and has a significant effect on the nature of the acid-rain formation process.

There are numerous mechanisms by which pollutants can attach to cloud and precipitation elements after the elements already exist, and thus in a manner that is consecutive with Step 1-2. These mechanisms are itemized in the following paragraphs; they are typically active for both aerosols and gases, although the relative importances and magnitudes vary widely with the state of the scavenged substance.

Diffusional attachment, as its name implies, results from diffusional migration of the pollutant through the air to the water surface. This process may be effective both in the case of suspended cloud elements and falling hydrometeors. It depends chiefly on the magnitude of the pollutant's molecular (or Brownian) diffusivity; and since diffusivity is inversely related to particle size, this mechanism becomes less important as pollutant elements become large. Diffusional attachment is of utmost importance for scavenging of gases and very small aerosol particles. For all practical purposes it can be ignored for aerosol particle sizes above a few tenths of a micrometer.

In concordance with Fick's law (Bird et al. 1960), diffusional transport to a water surface is dependent as well on the pollutant's concentration gradient in the vicinity of this surface. Thus if the cloud or precipitation element can accommodate the influx of pollutant readily, it will effectively depopulate the adjacent air, thus making a steep concentration gradient and encouraging further diffusion. If for some reason (e.g., particle "bounce off" or approach to solute saturation) the element cannot accommodate the pollutant supply, then further diffusion will be discouraged. If the cloud or precipitation element, through some sort of outgassing mechanism, supplies pollutant to the local air, then the concentration gradient will be reversed and diffusion will carry the pollutant away from the element.

Mixing processes inside of cloud or precipitation elements play an important role in determining the accommodation of gaseous species. If mixing is slow, for

example, it is likely that the element's outer layer will saturate with pollutant and thus inhibit further attachment processes. This is quite often a limiting factor in cases involving gas scavenging by ice crystals. Internal mixing occurs as a consequence of diffusion and fluid circulation and has been analyzed at length by Pruppacher and his co-workers (cf. Pruppacher and Klett 1978).

In general diffusional attachment processes are sufficiently well understood to allow their mathematical description with reasonable accuracy, and numerous references are available as guides for this purpose (e.g., Hales 1983, Pruppacher and Klett 1978, Slinn 1983).

Inertial attachment processes are directly dependent on the size of the scavenged particle and thus are unimportant for gaseous pollutants. In a somewhat general sense this class of processes depends on motions of pollution particles and scavenging elements relative to the surrounding air, which arise because both have finite volume and mass. The most important example of inertial attachment is the impaction of aerosols on falling hydrometeors. Here the hydrometeor (because of its mass and volume) falls by gravity, sweeping out a volume of space. Some of the aerosol particles (because of their mass) cannot move sufficiently rapidly with the flow field to avoid the hydrometeor and thus are impacted. In principle impaction could occur, even if the aerosol particles were point masses with zero volume. Assigning a volume to a particle further increases its chance of collision, simply on the basis of geometric effects. The inclusion of aerosol volume has been generally referred to in the past literature as accretion.

A second example of inertial attachment is turbulent collision. In this case the particles and scavenging elements, subjected to a turbulent field, collide because of dissimilar dynamic responses to velocity fluctuations in the local air. This scavenging mechanism is thought to be of secondary importance and has received comparatively little attention in the past literature, although some recent theoretical analyses have suggested it to be significant for specific drop-size/particle-size ranges.

While the mechanisms of diffusional and inertial attachment are efficient for capturing very fine and very coarse particles, respectively, a region of low efficiency should exist in the 0.1-5 μm range, where the mechanism is effective. This effect is shown schematically in Figure C.3-3. Because its importance to scavenging was first recognized by Greenfield (1957) it has become known

generally as the "Greenfield gap." Depending on circumstances there are several additional attachment mechanisms (including the two-stage nucleation-impaction mechanism mentioned earlier) that can serve to "fill" the Greenfield gap. Some of the more important of these are itemized in the following paragraphs.

Diffusiophoretic attachment to a capturing element can occur whenever the element grows via the condensation of water vapor. In effect the flux of condensing water vapor "sweeps" the surrounding aerosol particles to the element's surface. In a competitive cloud-element system where some droplets grow while others evaporate, diffusiophoresis can be a rather important secondary attachment mechanism. This is particularly true when the cloud contains mixtures of ice and liquid. Under such conditions the ice crystals have a pronounced tendency, owing to their lower equilibrium vapor pressure, to gain water at the expense of the droplets. Known as the Bergeron-Findeisen effect, this process is important in precipitation formation as well as in diffusiophoretic enhancement.

Thermophoretic attachment results from a temperature gradient in the direction of the capturing element. Here the element acts essentially as a miniature thermal precipitator. Warmer gas molecules on the outward side of the aerosol particle impart a proportionately larger amount of momentum, resulting in a driving force toward the capturing element.*

Thermophoresis depends directly on the temperature gradient in the vicinity of the capturing element. In cloud and precipitation systems local temperature gradients are caused most often by evaporation/condensation effects; thus thermophoresis is usually strongly associated with diffusiophoresis,† and in fact these two processes often tend to counteract each other.

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*One should note that the precise mechanisms of thermal transport differ radically, depending on particle size (cf. Cadle 1965).

†As noted by Slinn and Hales (1983) inappropriate treatment of this relationship has caused erroneous conclusions to be drawn in some of the past literature. The reader should be cognizant of this if more detailed pursuit is intended.

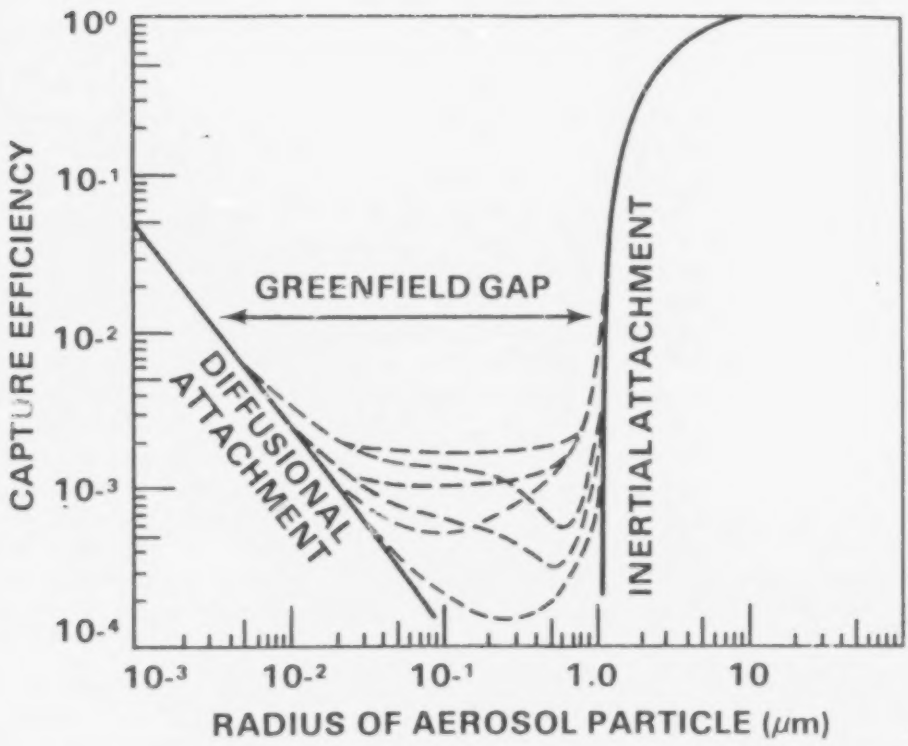

FIGURE C.3-4 Theoretical scavenging efficiency of a falling raindrop as a function of aerosol particle size. Adapted from Pruppacher and Klett (1978). Dashed lines correspond to contributions by electrical and phoretic effects under chosen humidity and raindrop-charge conditions (see original reference for details).

Phoretic processes are unimportant in the case of gaseous pollutants, owing to the overwhelming contributions of molecular diffusion. At present the theory of diffusiophoretic/thermophoretic particle attachment is at a state where reasonably quantitative assessments can be made for simple systems such as isolated droplets (Pruppacher and Klett 1978, Slinn and Hales 1971) (cf. Figure C.3.4). Rough estimates are possible also for more complex and interactive cloud/precipitation systems, but much remains to be done to bring our knowledge of this area to a really satisfactory state.

Electrical attachment of aerosol particles to cloud and precipitation elements has been the subject of continuing study over the past three decades. Understanding of this process is currently at a state where

relationships between aerosols and isolated drops can be quantified with reasonable accuracy (cf. Wang and Pruppacher 1977). In general, electrical charging of cloud and/or precipitation elements must be moderately high for electrical effects to become competitive with other capture phenomena, although such charging is certainly possible in the atmosphere--particularly in convective-storm situations. Understanding of electrical deposition in clouds of interacting drops is still at a relatively unsatisfactory stage of development.

As a conclusion to this discussion of attachment processes it is appropriate to note that, while the mechanisms have been presented here on an individual basis, they tend in actuality to proceed in a simultaneous and competitive manner. Insofar as atmospheric cleansing is concerned, this is a fortunate circumstance, because some mechanisms tend to be operative in physical situations where others are ineffective. Figure C.3-4 gives an excellent illustration of this point. Here theoretical attachment efficiencies appropriate to a 0.31-mm radius raindrop are presented for various electrical and relative-humidity conditions, demonstrating the capability of phoretic and electrical mechanisms to "bridge" the Greenfield gap. This simultaneous and competitive interaction of mechanisms serves to complicate profoundly the mathematics of the scavenging process and lends an additional degree of difficulty to the problem of scavenging calculations. This aspect will continue to emerge throughout this section, especially during the discussion of scavenging models.

3.1.4 Aqueous-Phase Reactions (Step 3-4)

Aqueous-phase conversion phenomena have been discussed in some detail in Appendix A and will not be examined further here except to note their general importance within the framework of the overall scavenging sequence. As observed previously in the context of Figure C.3-2, aqueous-phase reactions are not essential to the scavenging process. Depending on the pollutant material, however, these reactions often can have the effect of stabilizing the captured material within the condensed phase and thus enhancing the scavenging efficiency appreciably. There is much to be learned before this important aspect is brought to a satisfactory stage of understanding.

3.1.5 Deposition of Pollutant with Precipitation (Steps 3-5 and 4-5)

Although a variety of mechanisms exist (e.g., impaction of fog on vegetation), the predominant means for depositing pollutant-laden condensed water to the Earth's surface is simply gravitational sedimentation. Sedimentation rates depend on hydrometeor fall velocities, which depend in turn on hydrometeor size; thus the processes by which the pollutant-laden cloud droplets grow to precipitation elements emerge as major determining factors in this final stage of the scavenging sequence.

Once attached to condensed water, a pollutant molecule has several alternative pathways for action (Figure C.3-2). If the captured pollutant possesses some degree of volatility, it may desorb back into the gas phase. Reverse chemical reactions may occur. Evaporation of the condensed water may, in effect, "free" the pollutant to the surrounding gaseous atmosphere. This multitude of pathways results in an active competition for pollutant. If the precipitation stage of the scavenging sequence is to be effective, it must interact successfully within this competitive framework.

Besides competing actively for pollutants, the above interactions produce a vigorous competition for water. This parallel relationship between pollutant scavenging and water scavenging, apparent in some of the preceding discussion regarding attachment processes, can be drawn even more emphatically when considering precipitation processes. The following paragraphs provide a brief overview of some of the more important mechanisms in this regard.

Once initial nucleation has occurred, cloud particles may grow further by condensation of additional water vapor. Net condensation will occur to the surface of a cloud element whenever water vapor molecules can find a more favorable thermodynamic state in association with it; and because clouds contain varieties of makeup elements having different thermodynamic characteristics, a competition for water vapor usually exists. Such interactions are discussed at length in standard textbooks (Mason 1971, Pruppacher and Klett 1978). Slinn (1983) has developed a conceptual scavenging model in which condensational growth is an important rate-limiting step.

Thermodynamic affinity for water-vapor molecules depends on the cloud-element's size, its pollutant

burden, and its physical structure. These latter two factors often influence precipitation characteristics profoundly. In particular, the favored thermodynamic state of a water molecule in association with an ice crystal (as compared with a supercooled water droplet) results in rapid competitive growth of ice particles in mixed-phase clouds. This Bergeron-Findeisen process has been mentioned already in the context of diffusiophoretic and thermophoretic transport. Growth of large cloud elements via this process is the primary reason that ice-containing clouds tend to be so strongly effective as generators of precipitation water.

A further mechanism by which suspended cloud droplets can grow to form precipitation elements is coagulation. This process occurs via the collision of two or more cloud elements to form a new element containing the total mass (and pollutant burden)* of its predecessors. Coagulation occurs over size-distributed systems of cloud elements by a variety of physical mechanisms, and because of this it is a rather poorly understood and mathematically complex process. Comprehensive analyses of coagulation processes have been performed by Berry and Reinhardt (1974). Coagulation can be considered to be an important initiator of precipitation in single-phase clouds (water or ice). In mixed-phase clouds the Bergeron-Findeisen process can be expected to enhance the coagulation process by widening the droplet size distribution, as well as contributing to precipitation growth in a direct sense.

Once a moderate number of precipitation-sized elements have been generated, the process of accretion rapidly begins to dominate as a means for generating precipitation water. As noted previously, this process occurs by the "sweeping" action of large hydrometeors falling through the field of smaller elements, attaching them on the way. As was the case with coagulation, the accretion process tends to accumulate the pollutant burden of all collected elements.

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*Coagulation is often referred to as autoconversion in the cloud-physics literature. It is interesting to notice in this context that while coagulation tends to accumulate nucleated pollutants, the Bergeron-Findeisen process tends to re-liberate nucleated pollutants to the air.

Accretion can occur via drop-drop, drop-crystal, and crystal-crystal interactions. Drop-crystal interactions are particularly important in mixed-phase clouds; when supercooled droplets are accreted by falling ice crystals, the process is usually referred to as riming.

Although the above discussion has been confined primarily to deposition in conjunction with rain and snow, it should be emphasized that fog deposition* often is an important secondary process for conveying pollutants to the Earth's surface. Classification of fog-bound pollutant deposition is problematic for two major reasons. The first of these is that no sharp demarkation exists between "fog droplets" and "water-containing aerosols"; thus the choice of considering fog deposition as simply the dry deposition of wet particles, or the wet deposition of contaminated water, depends primarily on personal preference. Secondly, there is no real distinction between fog droplets and precipitation. Cloud physicists often find it convenient to categorize condensed atmospheric water into "precipitation" and "cloud" classifications, with the presumption that cloud water has a negligible sedimentation velocity. Such a classification is of limited use when considering fog deposition, however, owing to the fact that fog droplets do have significant gravitational fall speeds. A 50-μm-diameter fog droplet, for example, will fall at a rate of about 10 cm/s. This, combined with the fact that typical fogs and clouds contain droplet-size distributions ranging between 0 to 100 μm (cf. Pruppacher and Klett 1978), suggests that gravitational transport of fog droplets will indeed be a significant pollution-deposition pathway under appropriate circumstances.

In addition to purely gravitational transport, fog droplets have a strong tendency to impact on projected surfaces. The rates of fog impaction depend in a complex fashion on drop size, wind velocity, and geometry of the projected object. The common observations of rime-ice accumulation on alpine forests and on power-transmission lines give direct testimony to the effectiveness of this process.

Chemical deposition by fogs is directly proportional to fog-bound pollutant concentration, and this fact often acts to enhance substantially the pathway's overall

*A "fog" is (rather pragmatically) defined here as any cloud that is in the proximity of the Earth's surface.

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effectiveness. Owing to their proximity to the Earth's surface, fogs typically form in conjunction with high pollutant concentrations. Attaching particles and gases via the variety of mechanisms described in Section 3.1.3, the droplets typically accumulate extremely high burdens of material. It is not difficult to find evidence in support of this point. Scott and Laulainen (1979), for example, reported sulfate and nitrate concentrations approaching 500 μm/liter in water obtained near the bases of clouds over Michigan, while the SUNY group has reported (e.g., Falconer and Falconer 1980) numerous similar concentrations (as well as extremely low pH measurements) in clouds sampled at the Whiteface Mountain, New York, observatory.

Recently Waldman et al. (1982) have reported nitrate and sulfate concentrations in Los Angeles fogs ranging up to and beyond 5000 μm/liter. This compares with typical precipitation-borne concentrations of about 35 μm/liter for the northeastern United States.

Recently Lovett et al. (1982) have applied a simple impaction model to estimate fog-bound pollutant deposition to subalpine balsam fir forests and have concluded that chemical inputs via this mechanism exceed those by ordinary precipitation by 50-300 percent. This is undoubtedly an extreme case, and it would be more meaningful to possess a regional assessment indicating the general importance of fog deposition on an areal basis. This requires substantial effort however, involving climatological fogging analysis (cf. Court 1966) as well as numerous additional factors, and no really satisfactory evaluation of this type is currently available. Regardless of this it is appropriate to conclude that fog-deposition processes probably play an important, if secondary, role in pollutant delivery on a regional basis. In the future more effort should be addressed to this important research area.

3.1.6 Combined Processes and the Problem of Scavenging Calculations

The preceding discussion of individual steps in the scavenging sequence has been presented intentionally on a highly visual and nonmathematical basis, with appropriate references given for the reader interested in more detailed pursuit. Despite the qualitative nature of this presentation, however, it should be obvious that the most

direct and expedient approach to model development is first to formulate mathematical expressions corresponding to each of these steps and then to combine them in some sort of a model framework that describes the composite process. This subject will be examined in greater detail in Section 3.4, which is addressed specifically to scavenging models.

3.2 STORM SYSTEMS AND STORM CLIMATOLOGY*

3.2.1 Introduction

From the preceding discussion it is not difficult to imagine that scavenging rates and pathways will be dictated to a large extent by the basic nature of the particular storm causing the wet removal to occur. Storms containing water that is predominantly in the ice phase, for example, will provide little opportunity for attachment mechanisms associated with droplet nucleation, accretion, or phoretic processes. The abundance of liquid water and the temperature distribution in a given storm will have a direct bearing on the degree to which aqueous-phase chemistry can occur. Storms containing no ice phase whatsoever will be generally ineffective as generators of precipitation and thus will tend to inhibit the scavenging process. An interesting indication of the importance of storm type in this regard is presented in Figure C.3-23, which presents estimated scavenging efficiencies that vary extensively with storm classification. Different storm types differ profoundly with regard to inflow, internal mixing, vertical development, water-extraction efficiency, and cloud physics; and consequently it is appropriate at this point to consider briefly the major classes and climatologies of storm systems occurring over the continental United States.

Two major points should be stressed at the outset to this discussion. The first is the essential fact that all storms are initiated by a cooling of air, which leads to a condensation process. Such cooling may occur by the

*In the present text the term "storm" is intended to denote any system in which precipitation occurs. This definition thus encompasses all occurrences, ranging from mild precipitation conditions up to and through the major and cataclysmic events.

transport of sensible heat, such as when a comparatively warm, moist air parcel flows over a cold land surface. The dominant cooling mode for most storm systems, however, is expansion, which occurs via vertical motion of the air parcel to elevations of lower pressure. The second noteworthy point in this context is that the overwhelming majority of storm systems is strongly associated with fronts between one or more air masses. The primary reason for this fact, of course, is that thermodynamic perturbations and discontinuities associated with the frontal surfaces provide the opportunity for vertical motion (and thus expansion processes) to occur. This relationship is an essential component of storm-classification systems and will emerge repeatedly in the following discussion.

Overlaps in the characteristics of different storm types render a strict classification largely impossible. For practical purposes, however, it is convenient to segregate mid-latitude continental storms into two classes, which are usually described as being "convective" and "frontal" in nature. These two major categories then can be subdivided further as deemed expedient for the purpose at hand, although it should be noted that significant overlap among storm types occurs even at this major level of classification. Frontal storms, for example, often possess significant convective character in their basic composition, and true convective storms often occur as the consequence of fronts. Because of this the following discussion will utilize storm classification primarily as a descriptive aid and will not belabor taxonomical detail beyond this rather pragmatic end.

3.2.2 Frontal-Storm Systems

Much of what is understood today regarding mid-latitude frontal-storm systems stems from the pioneering work of the Norwegian meteorologist Bjerknes, who conducted a systematic survey of large numbers of storm systems and from this developed a conceptual model of frontal-storm development and behavior. Characterized schematically in Figure C.3-5, the Bjerknes model can be understood most easily by considering a cool northern airmass, separated from a warm southern air mass by an east-west front, as indicated in Figure C.3-5A. The progression of figures represents a typical result of the atmosphere's natural

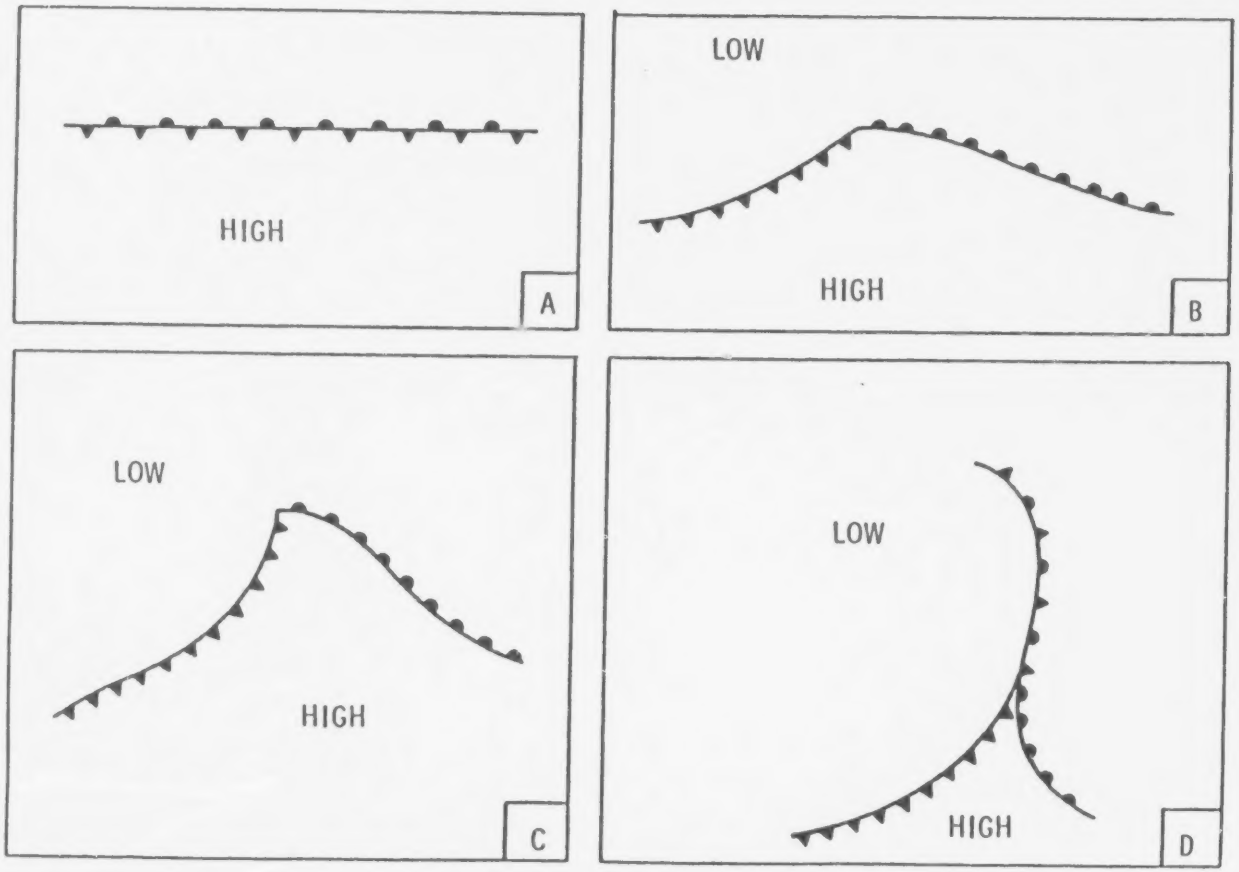


FIGURE C.3-5 Cyclonic storm development according to Bjerknes's conceptual model.

tendency to exchange heat from southern to northern latitudes. This is often referred to as a "tongue" of warm air intruding into the cold air mass. In the northern hemisphere this wave will tend to propagate in an easterly direction; thus the intrusion is bounded by two moving fronts--a warm front followed by a cold front, as shown in Figure C.3-5C.

Flows associated with the wave system occur in a manner such that a depression in atmospheric pressure occurs at the vertex of the warm-air intrusion, and as a consequence a general counterclockwise or "cyclonic" circulation pattern emerges. Because of this feature Bjerknes's conceptual model is often referred to as the "Bjerknes cyclone theory," and frontal storms associated with this pattern are termed "cyclonic" storms. A typical feature of storms of this type is the tendency for the cold front to overtake the warm front and, ultimately, to annihilate the wave. The "occluded" front created as a consequence of this behavior is shown schematically in Figure C.3-5D. In view of this birth-death sequence of the Bjerknes cyclone model, the progression depicted in Figure C.3-5 often has been termed the "life history" of a cyclone. Some idea of spatial scale and the general cyclonic flow pattern of a mature cyclone are given in Figure C.3-6. In viewing these indicated flow patterns, however, the reader should note carefully that considerable vertical structure exists in such systems, and marked deviations of the wind field with elevation are typical. In particular, one should take care not to confuse the indicated general circulation patterns with corresponding surface winds.

Although created from the limited observational base available during the early twentieth century, the fundamental precepts of the Bjerknes theory have proven valid even as more sophisticated observational and analytical facilities have become available. Certainly nonidealities and deviations from this model occur; but its general concepts have proven to be immensely valuable as a conceptual basis and as an idealized standard for the assessment of actual storm systems. Comprehensive descriptive and theoretical material pertaining to such systems is available in the classic text by Godske et al. (1957), and more elaborate and modern extentions are given in the periodical literature (e.g., Browning et al. 1973, Hobbs 1978).

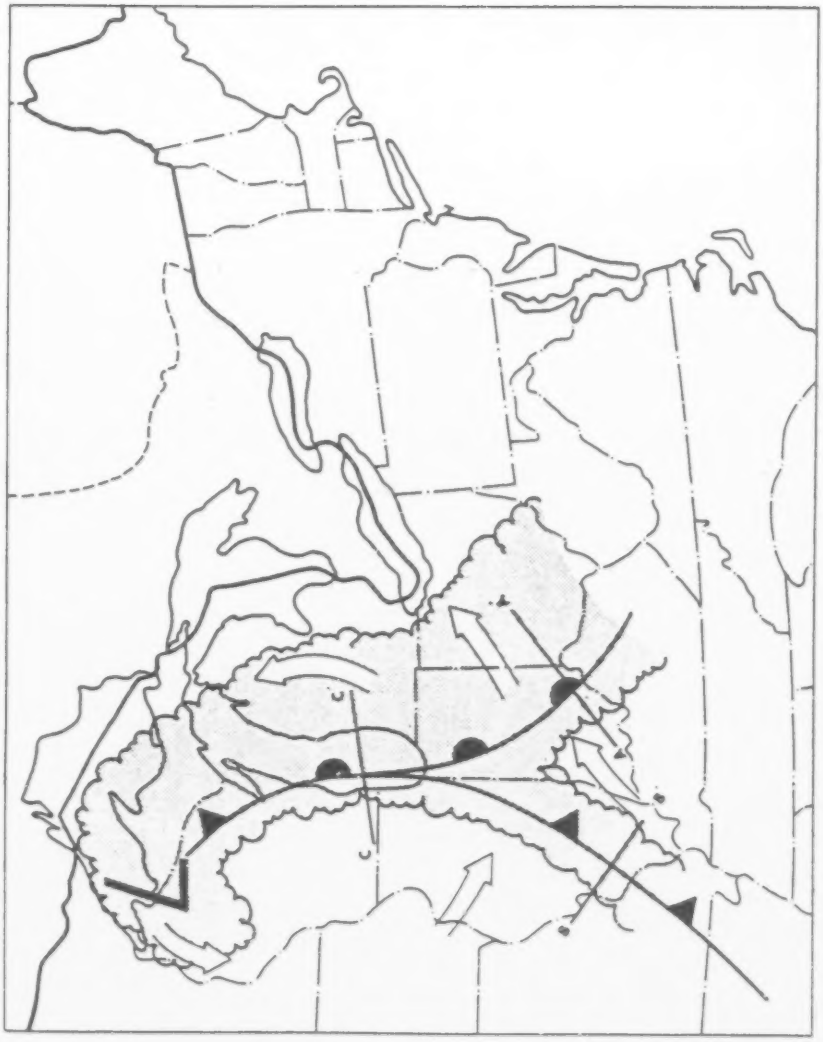


FIGURE C.3-6 General flow patterns in the vicinity of an idealized cyclonic storm system. Arrows denote general circulation patterns and should not be interpreted as surface winds.

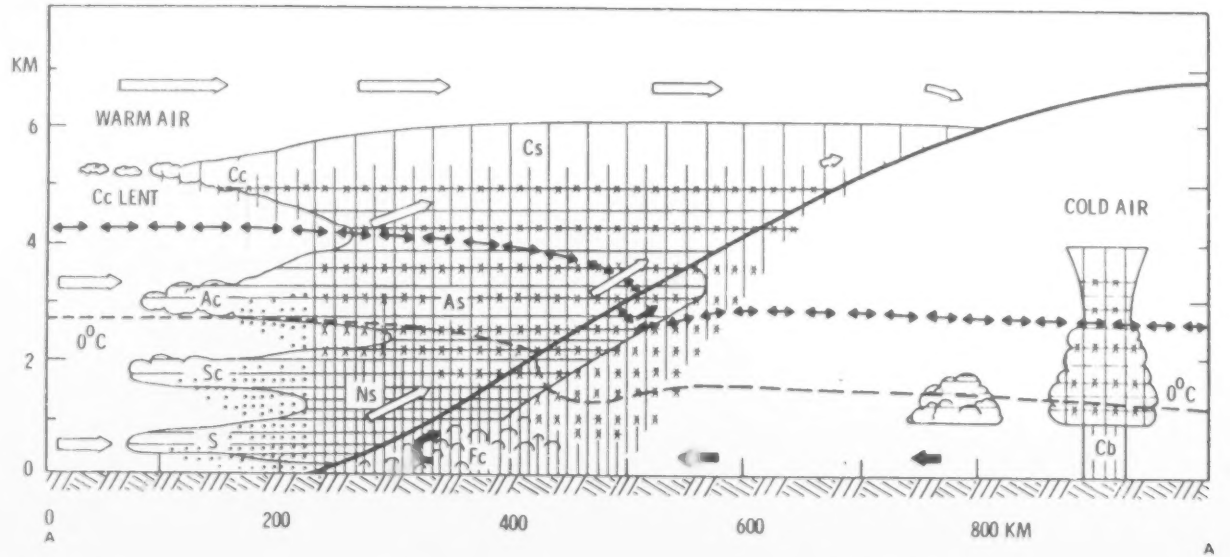
Warm-Front Storms

It is important to note that the plan views exhibited by Figure C.3-6 are gross simplications, since they do nothing to characterize the three-dimensional nature of the cyclonic system. If one were to construct a vertical cross section of the warm front (A-A' in Figure C.3-6), then typically one would observe an inclined frontal surface as shown in Figure C.3-7.* In this situation the presence of warm air aloft creates a relatively stable environment, which inhibits vertical mixing of air between the two air masses. The warm, moist air moves up over the cold air wedge, expanding, cooling, and ultimately forming clouds and precipitation. Typically the warm air supplying moisture for this purpose has been advected from deep within the southern air mass, carrying water vapor and pollutant over extensive distances. This transport trajectory has been aptly compared to a "conveyor belt" for moisture by Browning and his co-workers (1973). It is appropriate to note that this moisture conveyor belt is a conveyor belt for pollution as well.

Warm-front storms often are associated with long periods of continuous precipitation, although significant structure can exist within such systems. An important structural element in this regard is the occurrence of prefrontal rain bonds, which take the form of concentrated areas of precipitation imbedded within the major storm system. At present the factors contributing to rain-band formation are not totally understood, although mechanisms such as seeding from aloft by ice crystals and nonlinearities of the associated thermodynamic and flow processes undoubtedly contribute to a major extent.

Warm-front storms usually can be expected to be rather effective as scavengers of pollution originating from within the warm air mass, especially if temperatures in the feeder region are sufficiently high to allow the presence of liquid water and the nucleation-accretion process. Scavenging of pollutants from the underlying cold air mass usually will be less effective, owing to the relative scarcity of clouds and generally less definitive flows in this sector. Scavenging in both regions will of course depend on the physiochemical nature of the pollutant of interest and the microphysical attributes of the cloud system in general. Methods for

*See Table C.3-1 for definition of cloud abbreviations.



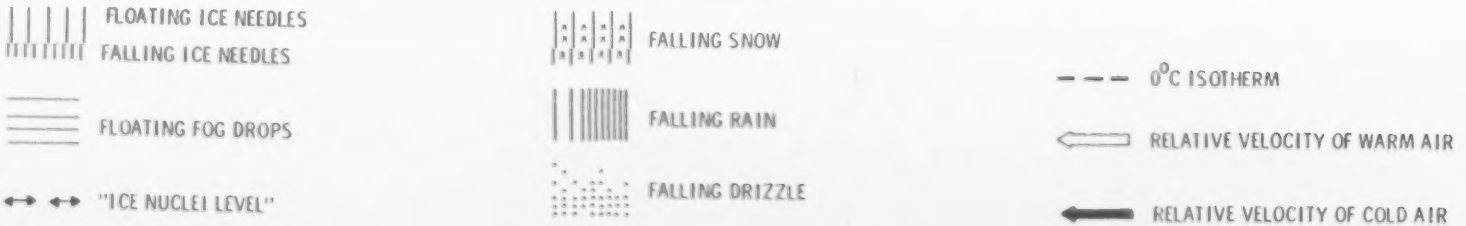


FIGURE C.3-7 Vertical cross section of a typical warm front (Section A-A' on Figure C.3-6). Adapted from Godske et al. (1957).

TABLE C.3-1 Summary of Cloud Types Appearing in Figures C.3-7-C.3-9

Type	Abbreviation
Cirrus	Ci
Cirrostratus	Cs
Cirrocumulus	Cc
Altostratus	As
Atlocumulus	Ac
Stratus	St
Stratocumulus	Sc
Nimbostratus	Ns
Cumulus	Cu
Cumulonimbus	Cb

estimating scavenging rates in such circumstances are discussed in Section 3.4.

Cold-Front Storms

A typical vertical cross section (B-B' in Figure C.3-6) of a cold-front storm is shown in Figure C.3-8. This differs substantially from the warm-front situation in the sense that, instead of flowing over the frontal surface, the warm air is forced ahead by the moving cold air mass. This action produces a more steeply inclined frontal surface, which, combined with the presence of low-elevation warm air, creates a relatively unstable situation leading to convective uplifting and the formation of clouds and precipitation.

Although discussed here in a frontal-storm context, this precold-front situation composes an important class of convective storms, which will be discussed in some detail later. Scavenging rates and efficiencies associated with such storm systems will again depend on the pollutant and the physical attributes of the particular cloud system involved.

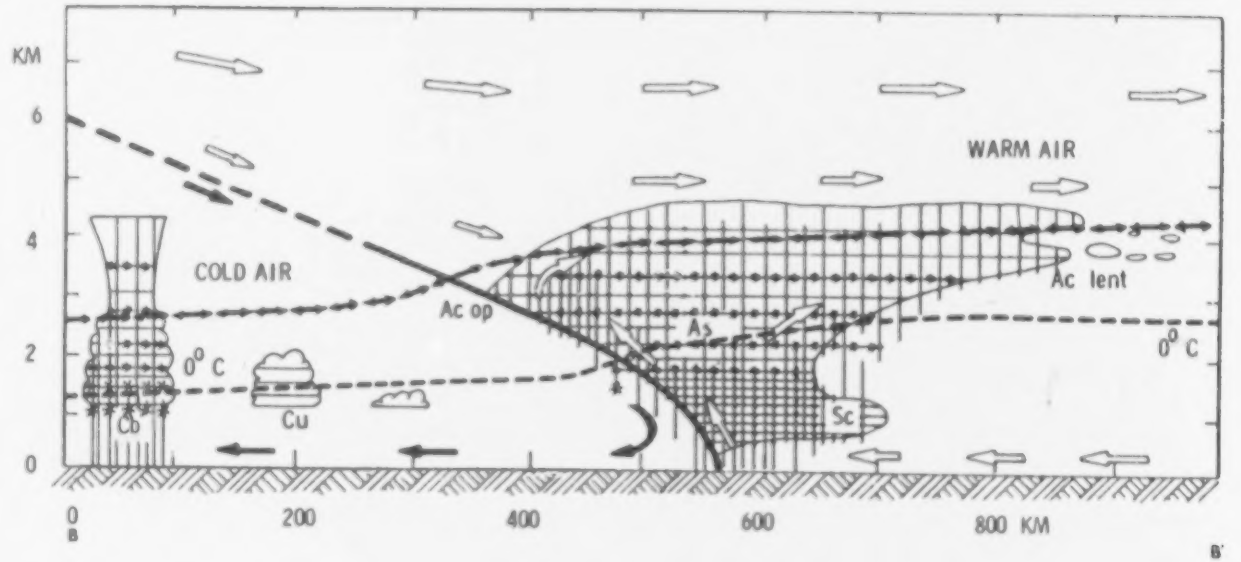


FIGURE C.3-8 Schematic vertical cross section of a typical cold front (Section B-B' in Figure C.3-6). Adapted grom Godske et al. (1957).

Occluded-Front Storms

Owing to the fact that occluded fronts are formed via merger of warm and cold fronts, it seems reasonable to expect that storms associated with occlusions should share characteristics of the respective elementary systems. Figure C.3-9, which shows a typical vertical cross section (Section C-C' on Figure C.3-6) of an occluded system demonstrates this point. Typically the easterly flow of warm air aloft maintains a relatively stable environment to the east of the occlusion, and clouds and precipitation occur in this region largely as a consequence of ascending flow from the south. Much more detailed accounts of occluded systems can be found in standard references such as the book by Godske et al. (1957).

3.2.3 Convective-Storm Systems

An idealized cross section of a typical convective storm is shown in Figure C.3-10. Such storms depend on atmospheric instabilities to induce the necessary vertical motions and concurrent cooling and condensation processes; and as such they are most likely to occur under warm, moist conditions where the energetics are most conducive to this process. Often convective storm systems occur as "clusters" of cells such as that shown in Figure C.3-10 and exhibit a marked tendency to exchange moisture and pollutant between cells; thus the flow dynamics and scavenging characteristics of such systems tend to be extremely complex.

Typically the moisture and pollutant input to a convective cell occurs primarily through the storm's updraft region (cf. Figure C.3-10), although entrainment from upper regions is possible as well. Dynamics of this process are such that violent updraft velocities often occur; these are capable of lifting entrained air, water vapor, and pollution to extremely high elevations (sometimes breaching the stratosphere). Along this course entrained pollutant is subjected to a large variety of environments and scavenging mechanisms; as will be noted in Section 3.4, convective storms tend to be highly effective scavengers of air pollution.

As was stated earlier, convective storms often are associated with frontal systems, although frontal influence is not absolutely necessary for their presence.

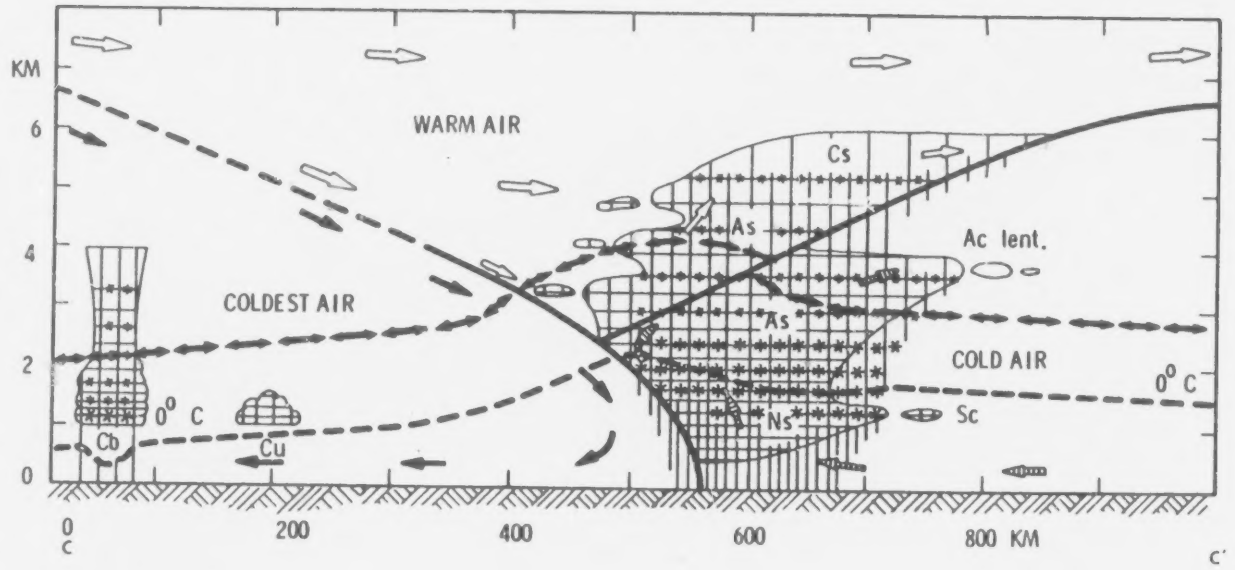


FIGURE C.3-9 Schematic vertical cross section of a typical occluded front (Section C-C'in Figure C.3-6). Adapted from Godske et al. (1957).

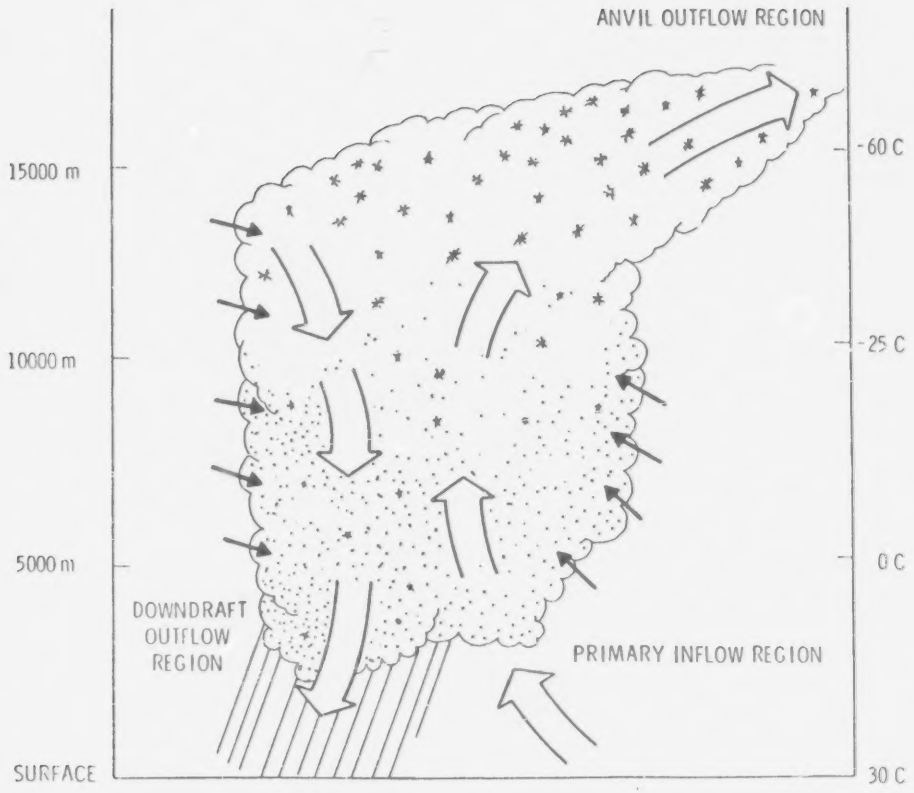


FIGURE C.3-10 Idealized cross section of an isolated convective storm.

An isolated air mass, for example, is totally capable of acquiring sufficient energy and water vapor to induce a convective disturbance on its own accord. Perturbations arising from fronts, however, often contribute to the creation of convective activity--if for no other reason than supplying a "trigger" to initiate convection in a conditionally unstable atmosphere.

3.2.4 Additional Storm Types: Nonideal Frontal Storms, Orographic Storms, and Lake-Effect Storms

As noted previously, the Bjerknes cyclone model represents something of an idealized concept, and numerous features can contribute to deviations from this "textbook" behavior. Orographic effects are highly important in this regard. Considering a cyclonic disturbance approaching the North American continent from across the

Pacific Ocean, for example, the frontal patterns typically lose much of their original identity after impacting with the mountainous regions of the west. In addition to the physical distortion of flow patterns, the lifting induced by the terrain encourages further precipitation, resulting in large spatial variability in rainfall patterns and pronounced local phenomena such as "rain shadows" and chinooks. Precipitation-formation and precipitation-scavenging processes associated with such systems tend to be highly complex.

Frontal systems often tend to reconstitute their structure after crossing the Rocky Mountains; but continental effects still impart a marked impact on their basic makeup. In the midwest-northeast region, for example, there is a tendency for the fronts to orient themselves in an east-west direction and become stationary for extended periods, often punctuated by several minor low-pressure areas. Even under relatively ideal conditions continental frontal storms tend to possess more convective flavor in their basic makeup than do their oceanic counterparts.

As indicated above, terrain-induced or "orographic" effects are usually most important in augmenting major storm systems, although relatively isolated orographic storms (such as oceanic "island-induced" storms) certainly do occur. Orographic effects obviously will tend to be most pronounced in regions where radical terrain changes occur; but even the small elevation changes typical of the Midwest can contribute significantly at times. Orographic effects also are suspected to influence storm behavior over substantial downwind distances. Lee waves from the Rocky Mountains, for example, have been suggested to trigger thunderstorm formation at extended distances.

Lake-effect storms are yet another example of a somewhat nonideal phenomenon that often is superimposed with more major meteorological patterns. Typically such storms occur during fall and early winter periods when land surfaces tend to be cooler than their adjoining water bodies. Considering an air parcel moving on an easterly course across Lake Michigan, for example, the warm lake surface tends to supply both heat and water vapor as it proceeds. As this parcel is advected across the downwind shore, however, two important things will occur. Firstly, the cold land mass will act to extract the heat from the air, and secondly the orographic lifting (of the order of a few tens of meters) will result in ascent, expansion, and further cooling. The

net result is a lake-effect storm. Such storms are capable of inducing highly variable precipitation patterns in specific areas around the Great Lakes region. Although confined largely to this portion of the United States, these storms are accountable for a majority of the snowfall accumulated in specific cities, such as Muskegon, Michigan, and Buffalo, New York. Some appreciation for the magnitude of this effect can be gained by looking at the climatological precipitation map given in Figure C.3-11.

3.2.5 Storm and Precipitation Climatology

The subject of storm climatology is exceedingly complex and will be discussed here only to the point necessary to describe some key attributes and indicate references for more detailed pursuit. Factors especially important in the context of precipitation scavenging are temporal and spatial precipitation patterns, storm-trajectory behavior, and storm-duration statistics. These will be discussed in order in the following paragraphs.

Precipitation Climatology

Figure C.3-12 provides climatological averages of monthly precipitation amounts at various stations throughout the United States. This figure was taken directly from the Climate Atlas of the United States (1968) and requires little elaboration at this point. It is interesting to note, however, that precipitation amounts do not vary radically throughout the year at most northeastern U.S. stations; this contrasts especially with the western and arid stations, whose seasonal variabilities tend to be pronounced. It should be noted as well that actual precipitation amounts for a given single month can vary appreciably from the climatological averages presented here.

Storm Tracks

Because of the difficulties noted previously with regard to precise classification or definition of storms, a really concise climatological summary of storm-pathway behavior is largely impossible. Some useful information can be generated, however, by observing the tracks of the

cyclonic (low-pressure) centers associated with major storm systems. Klein (1958), for example, has conducted a systematic survey of cyclonic centers in the northern hemisphere and from this has constructed monthly climatological maps of low-pressure tracks. Figure C.3-13, taken from the book by Haurwitz and Austin (1944), presents the combined results of the analyses by several previous authors. On the basis of the previous discussion it should be re-emphasized that, owing to the complex flow processes associated with cyclonic systems, one should not interpret the motion of these low-pressure centers as being identical with feeder trajectories for the storms themselves. Careful and skilled meteorological guidance is mandatory for the successful interpretation of such information in the context of source-receptor analyses.

Several additional points should be emphasized in the context of Figure C.3-13. Firstly it should be noted that this presents a long-term composite average and that marked deviations from this pattern can be expected to occur with season. Secondly the statistical variability of storm tracks is such that substantial departures from the long-term averages can be expected for any particular year. Finally, there is substantial evidence for longer-term shifts in average storm-track distributions (Zishka and Smith 1980); thus presentations (such as Figure C.3-13), which are based on historical data may vary considerably from storm patterns to be observed over the next 20 years. The implications of this with regard to long-term acid-deposition forecasting are obvious.

Additional features of cyclonic storm climatology can be found in standard climatological textbooks (e.g., Haurwitz and Austin 1944). Convective-storm climatology, which tends to be much more region-specific, can be evaluated from such references as well, although more recent weather modification programs such as METROMEX, NHRE, and HIPLEX have generated a considerable amount of new information in this area.

Storm-Duration Statistics

In the preparation of regional scavenging models it often is desirable to create some sort of statistical average of storm characteristics so that "average" wet-removal behavior can be defined. Although little activity has been devoted to this area until very recently, the

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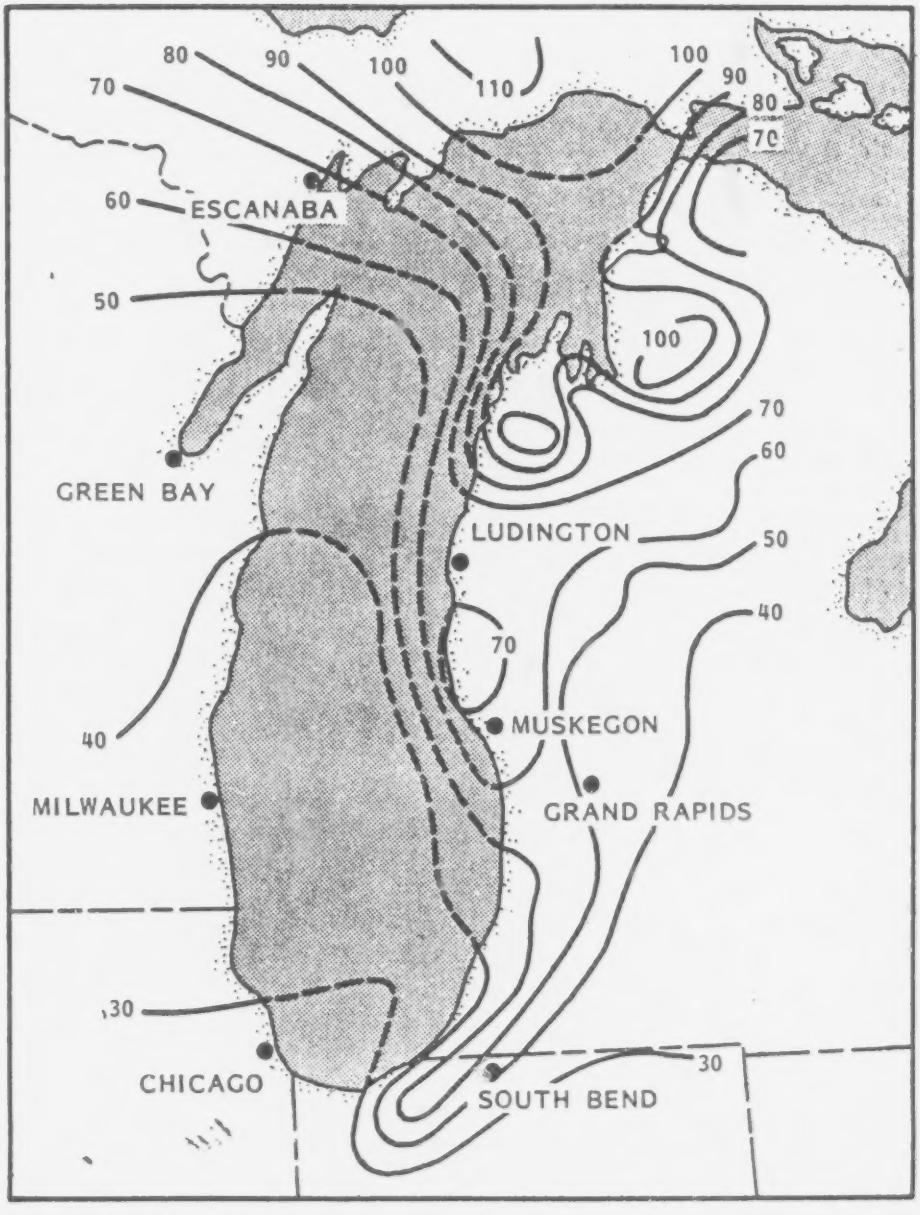
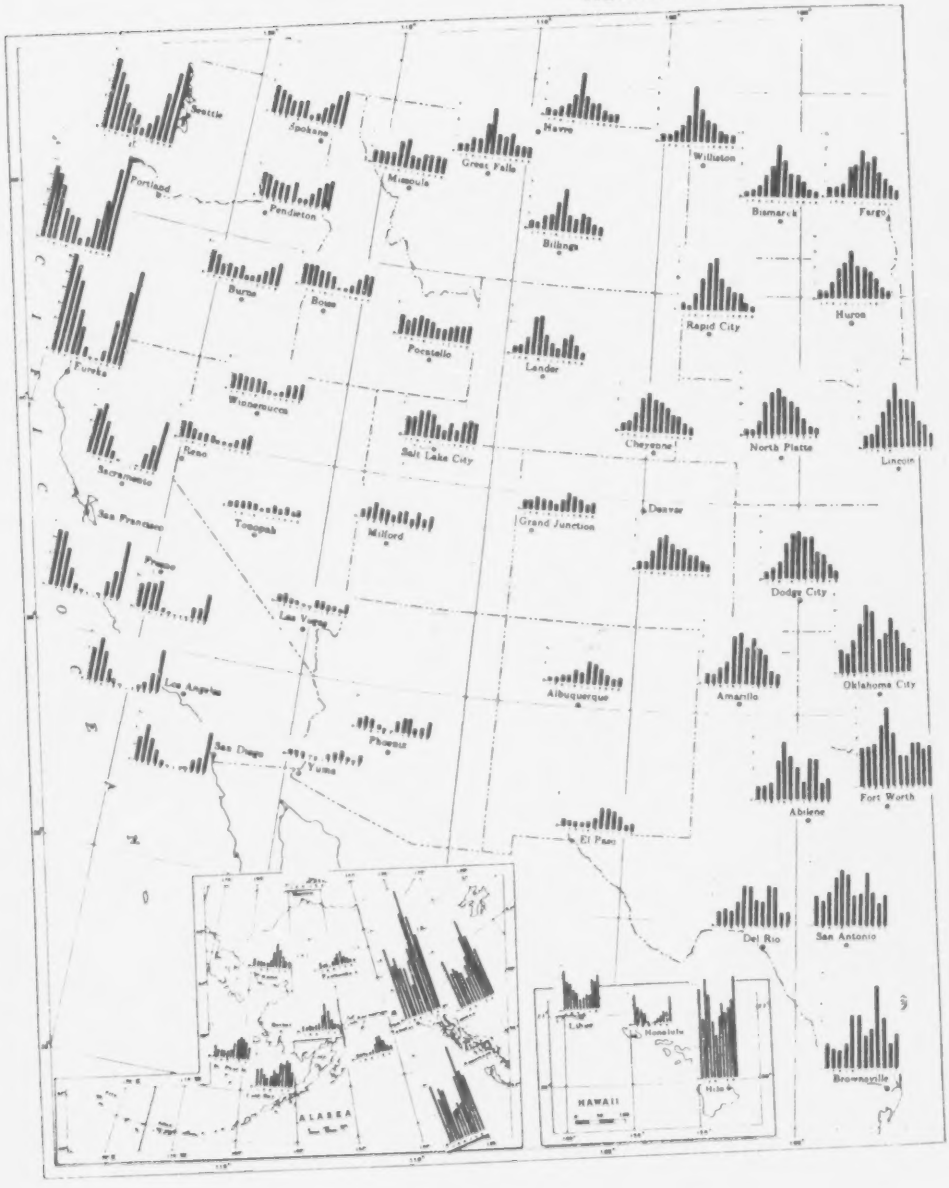


FIGURE C.3-11 Average annual snowfall pattern (inches) over Lake Michigan and environs. Adapted from Changnon (1968).

NORMAL MONTHLY TOTAL



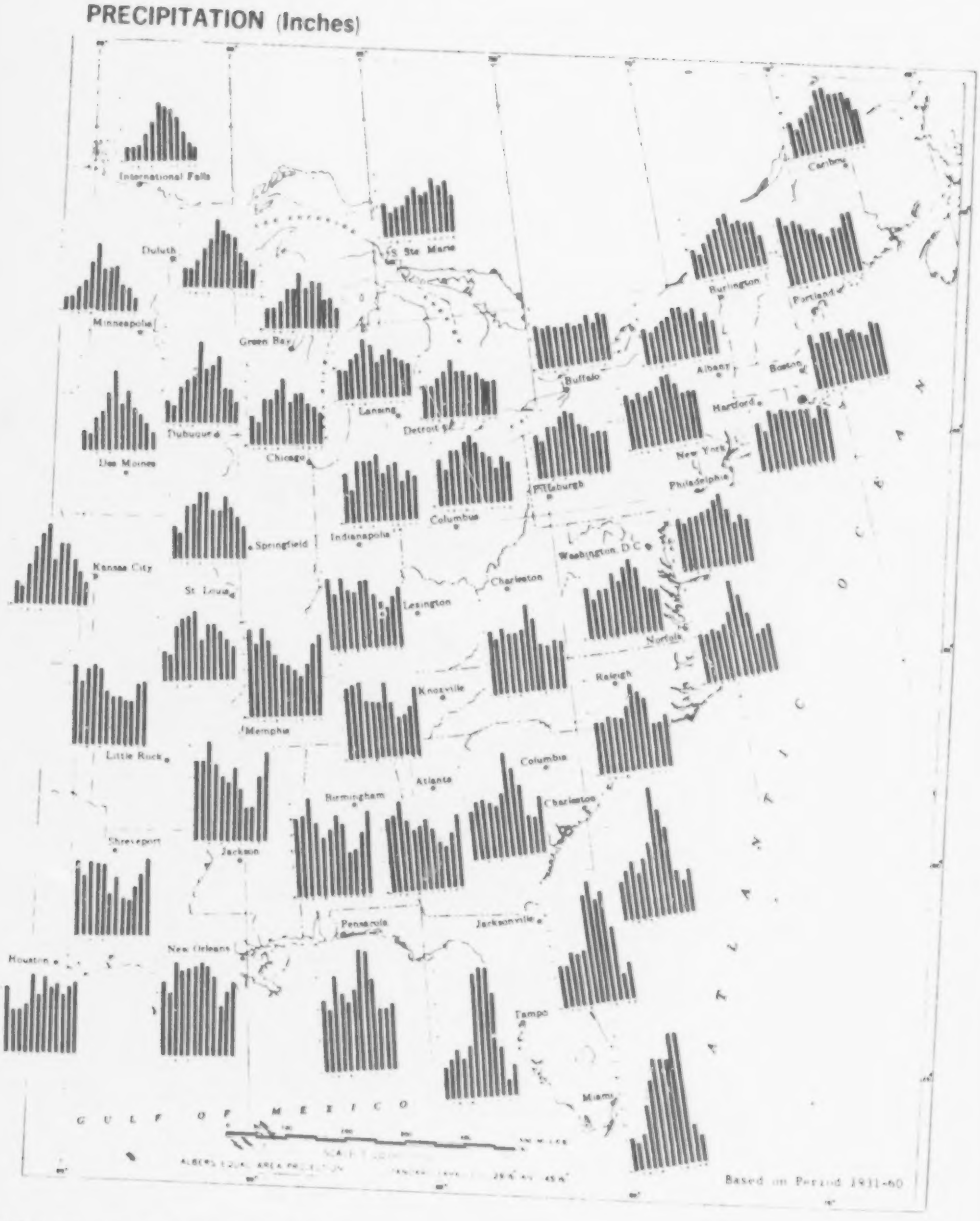


FIGURE C.3-12 Climatological summary of U.S. precipitation. From U.S. Climatological Atlas.

FIGURE C.3-13 Major climatological storm tracks for the North American Continent. Adapted from Haurwitz and Austin (1944). Dashed line denote tropical cyclone centers, and solid lines denote those of extratropical cyclones.

usefulness of such an approach to regional model development suggests accelerated effort during future years.

The analysis by Thorp and Scott (1982) provides an example of one such effort. These authors compiled data from hourly precipitation records from northeastern U.S. stations to obtain seasonally stratified duration statistics, which were expressed in terms of probability plots as shown in Figure C.3-14. As can be noted from these plots, "average" storm durations during summertime are significantly less than their wintertime counterparts, reflecting relative influences of short-term convective behavior. Some of the references given in Section 3.4 suggest potential modeling applications for these statistical summaries.

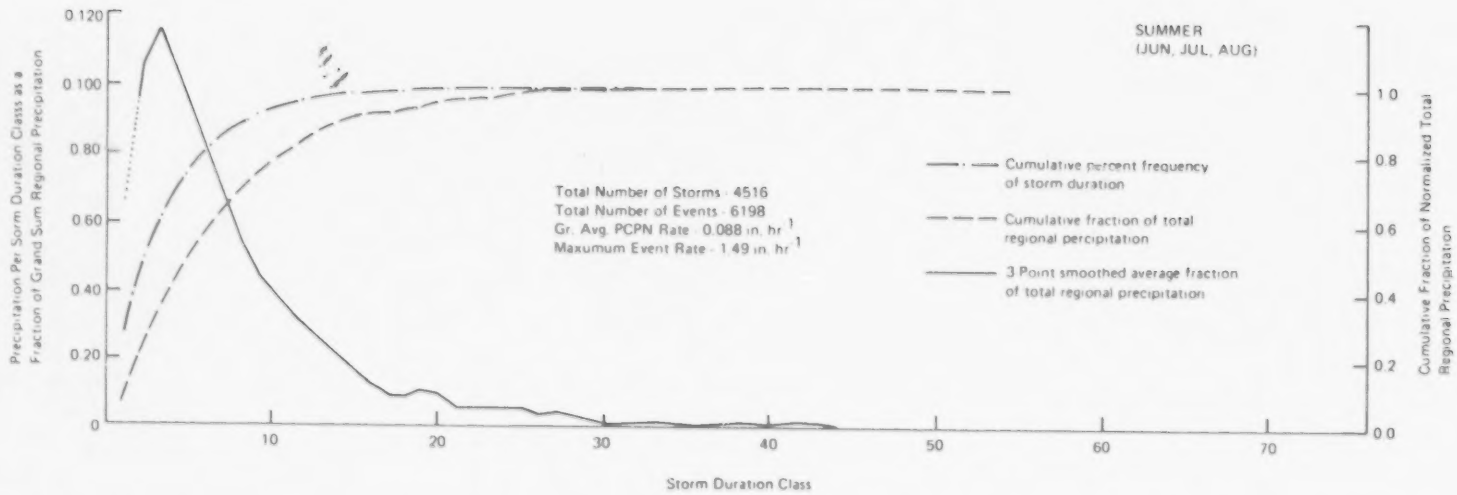


FIGURE C.3-14 Frequency statistics for storm duration, cumulative fraction of total regional precipitation, and smoothed average fraction of total regional precipitation. Summer (June, July, August) data from the MAP3S region. Frequency analysis allows up to 3 consecutive hours without precipitation to be counted as part of one storm.

3.3 SUMMARY OF PRECIPITATION-SCAVENGING FIELD INVESTIGATIONS

For the purposes of this document "field investigations" of precipitation-scavenging mechanisms will be differentiated from routine precipitation-chemistry network measurements, which are intended primarily for characterization purposes. There is of course a great deal of overlap between these two classes of measurements, and significant reciprocal benefit is generated as a consequence of each. There are some essential differences between the two, however, and it is convenient for present purposes to differentiate them accordingly.

The primary distinguishing feature of a scavenging field investigation is that the study usually is designed around the basis of some sort of conceptual or interpretive model(s) of scavenging behavior, which is tested on the basis of the field data. If the model predictions and data disagree, then some basic precepts of the model must be invalid, and additional mechanistic insights must be generated to rectify the situation. In the event that predictions and data agree, then this may be taken as evidence that the precepts may be correct. Regardless of whether positive or negative results are obtained (and assuming that the field study has been well designed and well interpreted), an advance in understanding has been achieved. The importance of such input cannot be overemphasized. Examples exist wherein field investigations have demonstrated then-accepted models to be in error by several orders of magnitude (e.g., Hales et al. 1971). Field studies have been essential in keeping the models "honest."

Field studies of precipitation scavenging began in earnest during the early 1950's to gain an understanding of radioactive fallout. Pioneering studies in this area were performed in England by Chamberlain (1953), which pertained to radioactive pollutant releases from point sources in anticipation of reactor accidents and related phenomena. These constituted the basis for the washout-coefficient approach to scavenging modeling (see Section 3.4). Other studies focused primarily on nuclear-detonation fallout and thus approached the scavenging problem from a more global point of view.

Following the English lead, nuclear-oriented studies were conducted by the United States, Canada, and the Soviet Union. These included studies of tracers as well as those of the radionuclides themselves, and although

some of this material still remains in the classified literature, it may be stated with certainty that most of what we know today regarding scavenging processes has been generated as a consequence of the nuclear era. The review "Scavenging in Perspective" by Fuquay (1970) presents a comprehensive account of this early stage of scavenging field studies.

During the late 1960s field-experiment emphasis shifted to more conventional pollutants, with the general recognition of precipitation scavenging's importance in preserving atmospheric quality and its potential adverse impacts of deposition on the Earth's surface ecosystem. Since that time a variety of large and small field studies have been conducted. These are summarized in Table C.3-2, which provides a logical classification in terms of source type, pollutant type, and geographical scale.

Although field studies have been focused strongly on quantitative aspects of precipitation scavenging, they have provided important qualitative information regarding acid precipitation processes as well. The ensemble of studies listed in Table C.3-2 presents a rather cohesive base of evidence in this regard; and although some conflicting results and uncertainties do exist, a generally coherent picture can be constructed in several important areas. Although there is considerable overlap of source-receptor distance scales among these studies, they tend to group rather conveniently into three classes of areal extent: 0-20 km, 0-200 km, and 0-2000 km. These classes shall be termed loosely as "local," "intermediate," and "regional" scales in the following discussion, where key qualitative features are illustrated by considering the fate of specific acidic-precipitation precursors (SO_x, NO_x, and HCl) as they are transported over these increasing scales of time and distance.

On a local scale (0-20 km) field studies have generally demonstrated the precipitation scavenging of sulfur and nitrogen oxides from conventional utility and smelting sources to be minimal. The virtual absence of excess nitrate or nitrite ion in precipitation samples collected beneath such plumes (Dana et al. 1976) provides strong evidence that direct uptake of primary nitric oxide and nitrogen dioxide by precipitation and cloud elements is a negligibly slow process.

Nonreactive scavenging of plumeborne sulfur dioxide is solubility dependent and tends also to be a rather

TABLE C.3-2 Summaries of Some Precipitation Scavenging Field Investigations

General Source Type	Specific Source Type	Selected References
Continuous point source	Tower releases of aerosols	Chamberlain (1953), Englemann (1965), Dana (1970)
	Tower releases of radioactive gases and simulated tracers	Chamberlain (1953), Engelmann et al. (1966)
	Tower releases of SO_2	Dana et al. (1972), Hales et al. (1973)
	Tower releases of tritiated water vapor	Dana et al. (1978)
	Tower releases of organic vapors	Lee et al. (1974)
	Power-plant plumes	Barrie and Kovalick (1978), Dana et al. (1973, 1976, 1982), Enger and Hogstrom (1979), Granat and Rodhe (1973), Granat and Soderland (1975), Hales et al. (1973), Hutchenson and Hall (1974), Radke et al. (1978)
	Smelter plumes	Kramer (1973), Larson et al. (1975)
"Instantaneous" and/or moving sources	Aircraft releases of rare-earth tracers	Changnon et al. (1981), Dingle et al. (1969), Gatz[a] (1977), Slinn (1973b), Young et al. (1976)
	Rocket releases of radioactive tracers	Burtsev et al. (1976), Shopauskas et al. (1969)
Urban sources	Uppsalla, Sweden	Hogstrom (1974)
	St. Louis, Mo.	Hales and Dana (1979a), Hales et al. (1979)
	Los Angeles, Calif.	Morgan and Liljestrand (1980)
General and regional sources	Regional pollution flowing into lake-effect storms	Scott (1981)
	Regional pollution in the eastern U.S. and Canada	Easter (1982), MAP3S/RAINE (1981)
	Regional aerosol loadings at a specific receptor point	Graedel and Franey (1977), Peters et al. (1978)
Global and stratospheric sources	Cosmogenic radionuclides	Young et al. (1973)
	Nuclear fallout	Numerous studies: see Fuquay (1970)

[a]The reference by Gatz provides a comprehensive list of past tracer studies of precipitation scavenging.

inefficient process, although it is definitely detectable in field studies conducted in relatively clean environments (Dana et al. 1973, 1976; Hales et al. 1973). This phenomenon, which is suppressed under conditions involving high rain acidity, is relatively well understood at present (Drewes and Hales, 1982).

Nonreactive scavenging of sulfate aerosol can be an efficient removal process. The preponderance of relevant field tests of Table C.3-2, however, has demonstrated that wet deposition of sulfate from local power-plant and smelter plumes occurs rather slowly. This is undoubtedly a consequence of the small amounts of primary sulfate available for scavenging under such circumstances.

Field tests conducted under situations wherein sulfur trioxide was intentionally injected into the stack of a coal-fired power plant (Dana and Glover 1975) show correspondingly high sulfate scavenging rates, and it has been suggested that under certain operating conditions some types of power plants (especially oil-fired units) will produce sufficient primary sulfate to account for appreciable local deposition. To date, however, there has been no really strong field evidence in support of this point. Hogstrom et al. (1974) reported the observation of substantial sulfate scavenging from the local plume of an oil-fired power plant in Sweden, but these results are rather dependent on the interpretation of background contributions. Granat and Soderlund (1975) performed a similar investigation in the vicinity of a second Swedish oil-fired plant and found a comparatively small scavenging rate.

Reactive scavenging of plumeborne sulfur dioxide to form rainborne sulfate is difficult to differentiate from primary sulfate removal. The previously noted findings of low excess sulfate in below-plume rain samples, however, suggests that neither process is particularly effective in near-source plume depletion.

The scavenging of hydrochloric acid to produce chloride and hydrogen ions in precipitation will most certainly be a highly effective process, depending on the quantities of hydrochloric acid available. Considerable theoretical and laboratory work has been conducted in this area for Space Shuttle impact assessment, and there are limited data suggesting that hydrogen chloride is scavenged in measurable amounts from power-plant plumes (Dana et al. 1982).

With the exception of studies conducted under rather clean ambient conditions (e.g., Dana et al. 1973, 1976)

the influence of background contributions has made the interpretation of plume scavenging a difficult task. Typically the sulfate and nitrate concentrations in precipitation collected adjacent to the plume are quite variable, and subtracting this influence to determine source contributions involves substantial levels of uncertainty. This difficulty of "source attribution" at the local scale is compounded appreciably as greater scales of time and distance are considered.

On a more intermediate scale (0-200 km) an enhancement of sulfate and nitrate precipitation scavenging seems to occur, presumably because the precursors have had more opportunity to dilute and to react under these circumstances. Hogstrom (1974) reported substantial scavenging rates of sulfur compounds using an extended network of samplers in the vicinity of Uppsala, Sweden. Hales and Dana (1979a,b) observed summertime convective storms to remove appreciable fractions of urban NO_x and SO_x burdens in the vicinity of St. Louis, Missouri. Although both of these studies were subject to the usual uncertainties with regard to background contributions, there is little doubt about their general conclusions of significant scavenging under such circumstances.

On a regional scale (0-2000 km) there are relatively few data from intensive field experiments. Precipitation-chemistry network data are of some utility in this regard, however, and several analyses have applied these measurements to specific ends. One result of these analyses is the suggestion that, in the northeastern quadrant of the United States, roughly one third of the emitted NO_x and SO_x is removed by wet processes (Galloway and Whelpdale 1980). Network data for the northeast (MAP3S/RAINE, 1982) show also that the molar wet delivery rates of NO_x and SO_x are roughly equivalent. Combining this result with regional emission inventories suggests that nitrogen compounds begin to wet deposit with a significantly enhanced efficiency as distance scales become regional in extent.

The above changes in behavior with increasing scale seem to be a logical consequence of current understanding regarding the atmospheric chemistry of SO_x and NO_x. On local scales neither is scavenged very effectively owing to the chemical makeup of the primary emissions. On intermediate scales both groups have had some opportunity to react into more readily scavengable substances. Depending on ambient conditions, the nitrogen oxides will have participated to some extent in initial photolysis

reactions and proceeded on to form scavengable products such as nitric acid, peroxyacetyl nitrate, and nitrate aerosol. Sulfur dioxide also will have reacted homogeneously to a limited extent; more importantly, however, this compound will have diluted to levels where limited reactants (and possibly catalysts) will facilitate its oxidation in the aqueous phase. On a regional scale this progression continues with the relative acceleration of NO_x scavenging.

Present field-study indications that NO_x scavenging may occur primarily through the attachment of gas-phase reaction products, while the scavenging of SO_x may depend much more heavily on aqueous-phase oxidation processes are also reflected in precipitation-chemistry data. A possible consequence of this difference in mechanisms is illustrated in Figure C.3-15, which is a time series of daily precipitation-chemistry measurements for a northeastern U.S. site. The decidedly periodic* behavior of sulfate-ion concentrations in contrast to the largely disorganized behavior of nitrate-ion concentrations has been suggested to occur as a consequence of an aqueous-phase oxidation of sulfur dioxide, which proceeds more rapidly during summer months. Whatever the cause, it is readily apparent from this figure that scavenging mechanisms for these two species differ appreciably.

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*One should note in Figure C.3-16 that the periodic functions are fit to the total data, whereas the linear regressions are fit only for the period January 1, 1977-December 31, 1979; thus the cyclic functions are not exactly symmetric about the linear regression curves. Some idea of statistical improvement in fit may be obtained using the expression

$$\hat{r}^2 = \frac{\sigma^2_{\text{linear regression}} - \sigma^2_{\text{periodic fit}}}{\sigma^2_{\text{linear regression}}},$$

where the σ^2's pertain to variances of the data points over the three and one-half period. For sulfate in Figure C.3-16, r^2 equals 0.22, indicating a significant reduction in variance; the corresponding r^2 value for nitrate is 0.01, suggesting that no significant annual periodicity exists in this case.

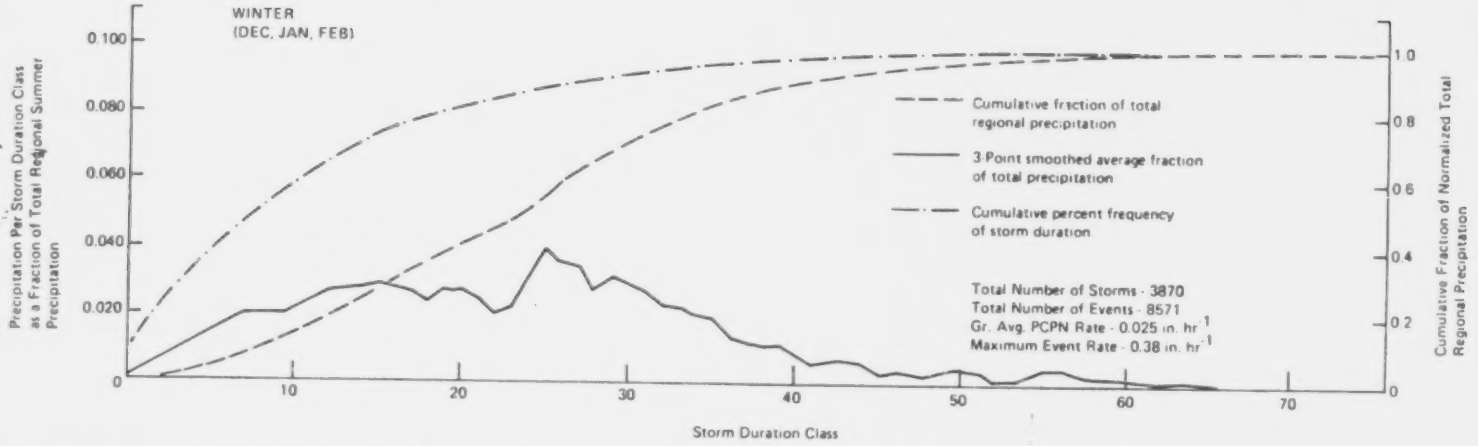

FIGURE C.3-15 Frequency statistics for storm duration, cumulative fraction of total regional precipitation, and smoothed average fraction of total regional precipitation. Winter (December, January, February) data from the MAP3S region. The frequency analysis allows up to 6 consecutive hours without precipitation to be counted as part of one storm.

As noted above, most past field experiments have experienced difficulty in resolving precisely which source(s) of pollution has been responsible for material wet-deposited at sampled receptor sites, and this problem is typically amplified as time and distance scales increase. Source attribution is particularly uncertain on a regional scale, and the basic data obtainable from standard precipitation-chemistry networks are of limited help in this regard. Combined with the lack of data from well-designed regional field studies, this aspect poses one of the most important and uncertain questions facing the acid deposition issue at present.

As a consequence of this need, a major regional field experiment has recently been designed and conducted in the northeastern United States (Easter 1982, MAP3S/RAINE 1981). Known as the Oxidation and Scavenging Characteristics of April Rains (OSCAR) study, this field experiment was based on the concept of characterizing, as completely as possible, the dynamical and chemical features of major cyclonic storm systems as they traverse the continent. Specific objectives were as follows:

1. To assess spatial and temporal variability of precipitation chemistry in cyclonic storm systems and to test the adequacy of existing networks to characterize this variability;
2. To provide a comprehensive, high-resolution data base for prognostic, regional deposition-model development;

and

3. To develop increased understanding of the transport, dynamical, and physicochemical mechanisms that combine to make up the composite wet-removal process and to identify source areas responsible for deposition at receptor sites.

The data collected and assembled by the OSCAR project are summarized in Table C.3-3. These are being made available to the general user community in a computerized data base.

A general layout of the OSCAR precipitation chemistry network is shown in Figure C.3-16. The points and triangles on this map represent locations of sequential precipitation-chemistry stations on an "intermediate-density" network, and the open square, overlapping Indiana and Ohio, depicts a concentrated network of 47 additional sites. Specific design criteria for this

TABLE C.3-3 Summary of Data Collected for the OSCAR Data Base

Meteorological Data

- North American standard 12-h upper-air observations (rawinsondes)
- OSCAR special rawinsonde data
- North American 3-h standard surface observations
- North American hourly precipitation amount data
- Trajectory forecast data (Limited Fine Mesh and Global Spectral Models)
- Gridded forecast data (Limited Fine Mesh Model)
- Satellite observations

Precipitation-Chemistry Data

- OSCAR network: Sequential measurements of rainfall, field pH, laboratory pH, conductivity, NO_3^-, NO_2^-, $SO_4^=$, $SO_3^=$, Cl^-, NH_4^+, Ca^{++}, Mg^{++}, K^+, Na^+, Al^{+++}, PO_4^X, total Pb
- Additional networks: Time-averaged data as available from sources such as NADP, CANSAP, CCIW, and APN
- Special rainborne H_2O_2 measurements

Aircraft Data

- Trace gases: O_3, NO/NO_x, SO_2, HNO_3, NH_3
- Aerosol parameters: scattering coefficient (b_{scat}), Aitken nuclei, aerosol sulfur, sulfate size distribution, aerosol size distribution, aerosol acidity
- Cloud water chemistry: NO_3^-, NO_2^-, $SO_4^=$, $SO_3^=$, pH, NH_4^+, conductivity, Cl^-, Ca^{++}, Mg^{++}, K^+, Na^+, total Pb
- Meteorological parameters: Temperature, humidity, liquid-water content, wind speed and direction, cloud droplet size distribution
- Position parameters: Latitude, longitude, altitude, time

Surface Air Chemistry Data

- OSCAR SAC site (Fort Wayne 40° 49.8' N, 85° 27.6' W): H_2O_2, peroxyacetyl nitrate, sulfur aerosol size distribution, NH_3, SO_2, $SO_4^=$, O_3, NO/NO_x, HNO_3, aerosol composition versus particle size, aerosol acidity
- Selected air -uality data from specific surface monitoring sites throughout eastern North America

Emissions

- MAP3S/RAINE standard inventory

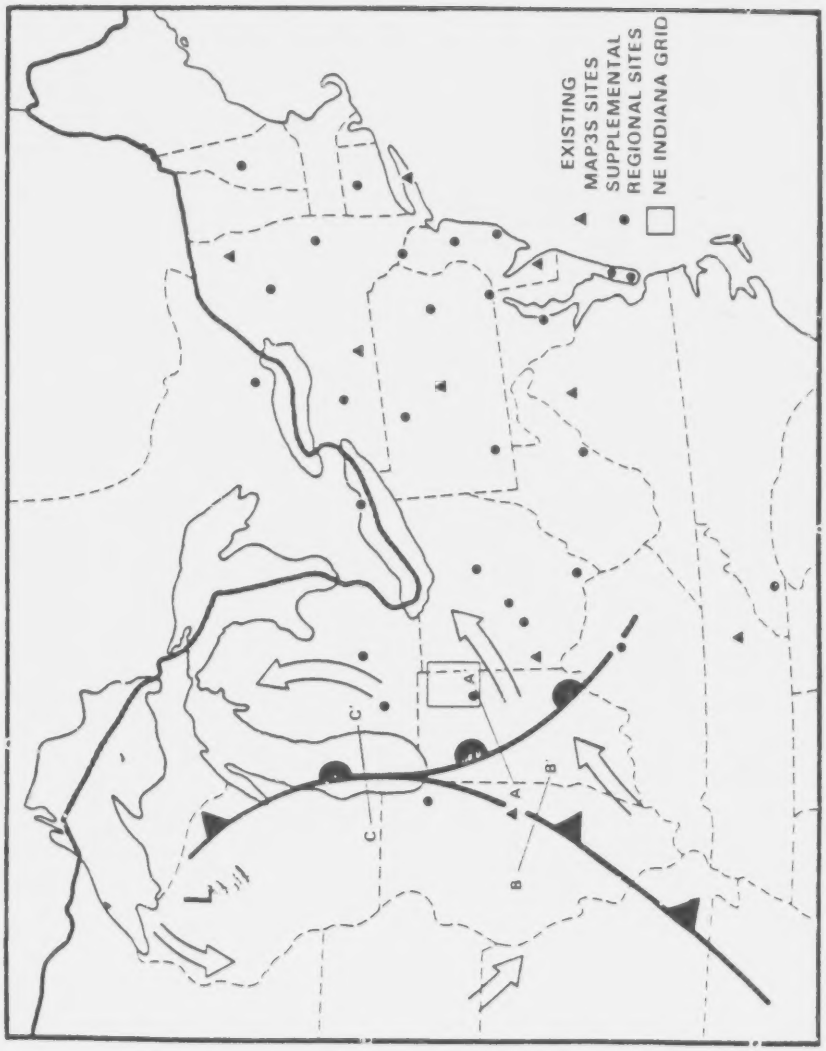

FIGURE C.3-16 General layout of OSCAR sequential precipitation chemistry network, showing hypothetical "design-basis" cyclonic system.

configuration are discussed in the supporting literature (MAP3S/RAINE 1982).

The OSCAR data set is currently under intensive analysis, and only preliminary results are available. It is of interest to consider some of these results at this point, however, to evaluate the potential future utility of this material. One early result, presented by Raynor (1981), is primarily of qualitative interest. These are the first-sample/last-sample pH data obtained by the sequential rain samplers for individual storms and are typified by the plots shown in Figures C.3-17 and C.3-18. It is interesting to note that Figure C.3-17 is strongly reminiscent of annual- or multiyear-average plots for the northeastern United States in the sense that it shows the familiar acid "core" region centered upon Pennsylvania.* The final-sample distribution in Figure C.3-18 is quite different. Besides indicating a much cleaner sample set, very little structure exists in this final distribution. This relative cleanliness of late-storm precipitation is consistent with the general OSCAR finding that most of the pollutant is scavenged comparatively early in a storm's life cycle (Easter and Hales 1983a).

Substantial source-receptor analysis is currently being conducted in conjunction with the Indiana-Ohio concentrated network. One early analysis, conducted for the April 22-24, 1981, storm, is presented in Figure C.3-19. Backtrajectories of this type are currently being combined in diagnostic scavenging models with aircraft and surface data to evaluate source-receptor relationships in greater detail (Easter and Hales 1983a,b).

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*It should be noted in this context that field studies having higher spatial resolution (e.g., Hales and Dana, 1979b, Semonin 1976) indicate that significant fine structure typically exists in spatial pH distributions. Much of this fine structure can be expected to be hidden within the relatively coarse sampling mesh shown in Figures C.3-18 and C.3-19.

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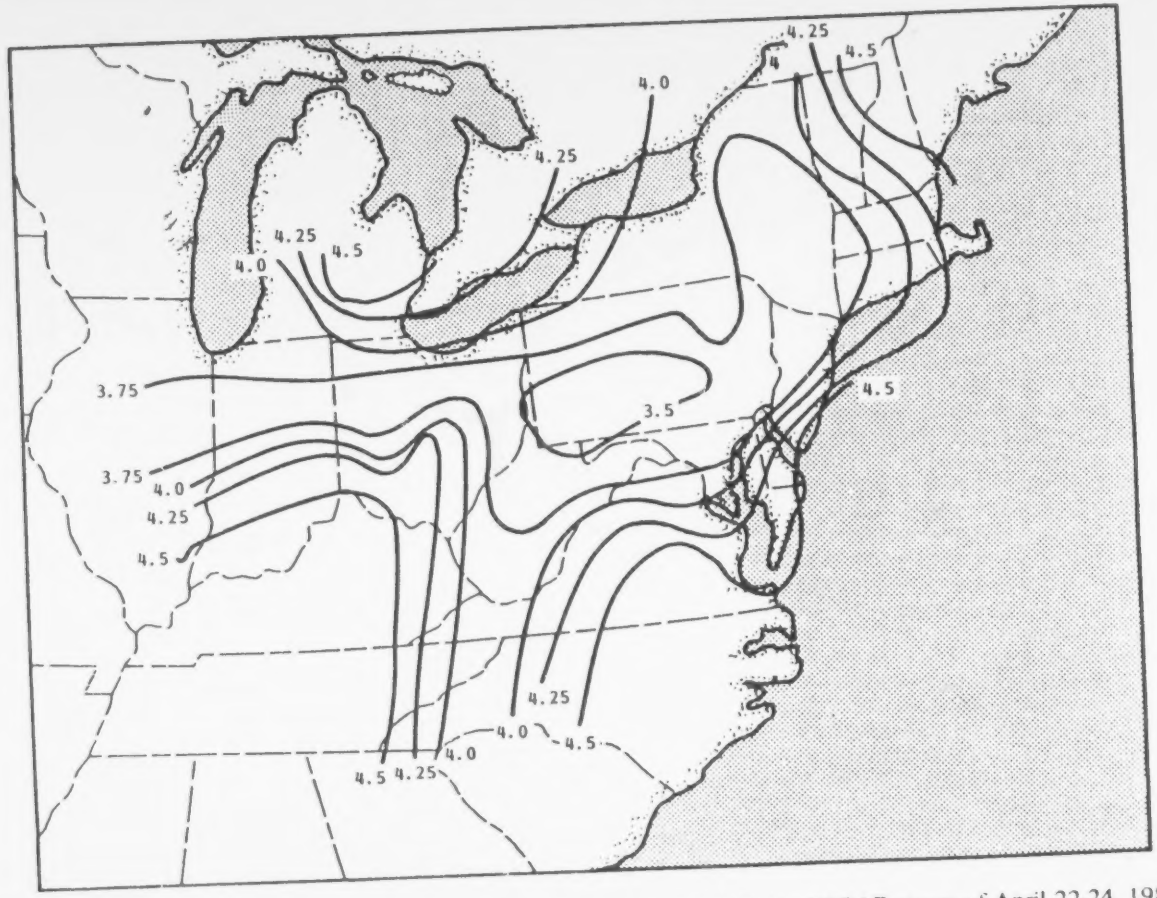

FIGURE C.3-17 Distribution of pH for initial precipitation sampled during OSCAR storm of April 22-24, 1981.

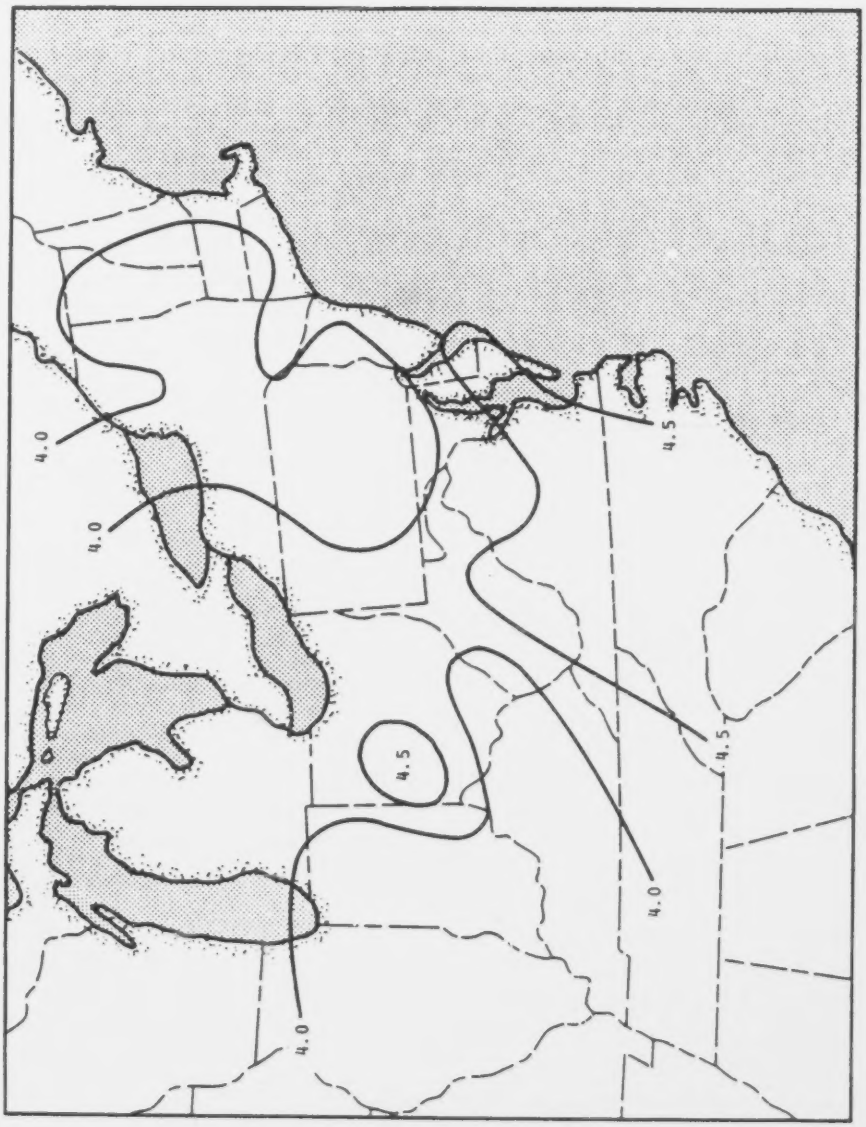

FIGURE C.3-18 Distribution of pH for final precipitation sampled during OSCAR storm of April 22-24, 1981.

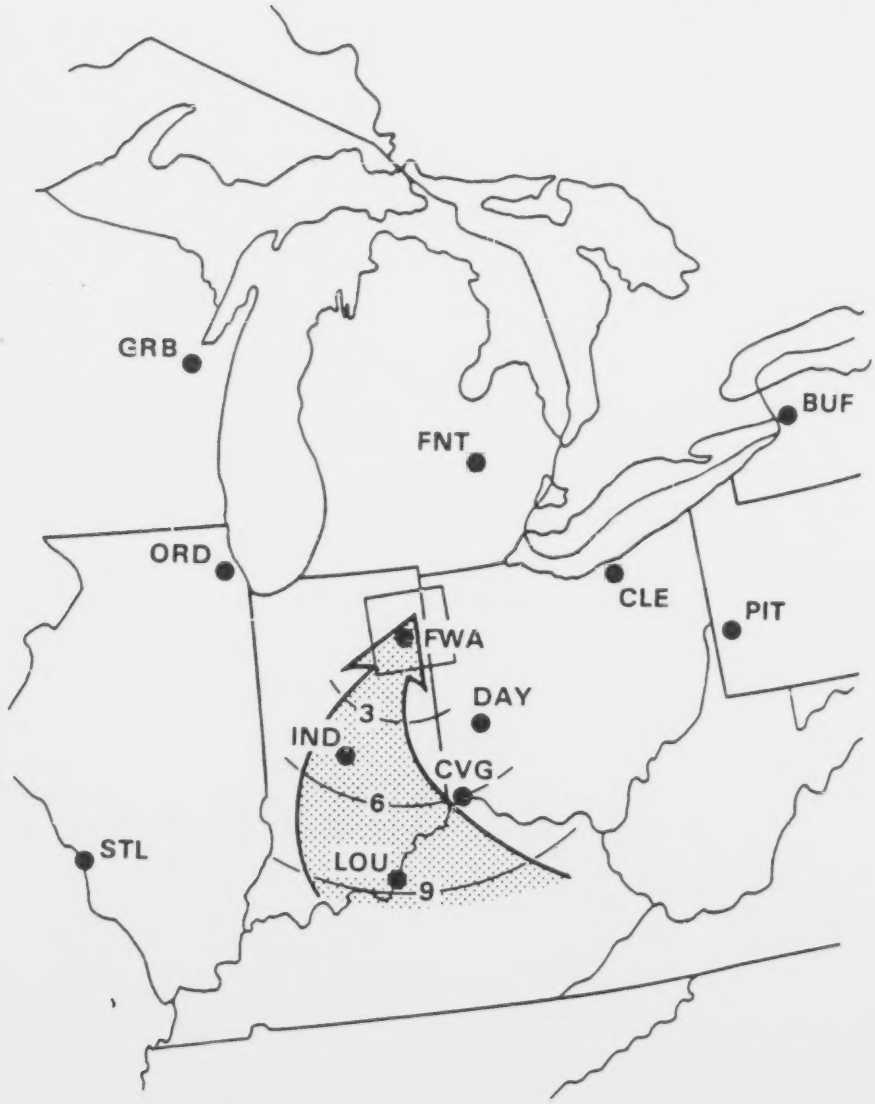

FIGURE C.3-19 Loci of points contributing pollution to the high-density network near 1400 EST on April 22, 1981. Contour intervals 3, 6, 9 represent travel times in hours from source regions. The large arrow represents the likely path of air originating from points 9 hours upwind of the receptors.

3.4 PREDICTIVE AND INTERPRETIVE MODELS OF SCAVENGING

3.4.1 Introduction

A precipitation-scavenging model can be defined as any conceptualization of individual or composite processes of Figure C.3-2, in a manner that allows their expression in mathematical form. Often such models take the form of submodels or "modules" within a larger calculational framework, such as a composite regional pollution code. When considered in a modular sense the lines connecting the boxes of Figure C.3-2 can be considered as channels for information exchange within the overall framework, whereas the boxes (or clusters of boxes) can be identified with the modules themselves. Scavenging models are currently in a rapidly evolving state, and a profusion of associated computer codes and computational formulas is currently available. Indeed, one of the major problems in precipitation-scavenging assessment is determining precisely which model to select from the large number of available candidates. A major aim of the present subsection is to guide the reader in this pursuit.

There are a number of potential uses for precipitation-scavenging models, and the intended use will to a large extent determine which model should be employed. Some of the more important potential uses are itemized as follows:

- Prediction of the impact on precipitation chemistry of proposed new sources, source modifications, and alternate emission-control strategies;
- Prediction of long-range trends in precipitation chemistry;
- Estimation of the relative contributions of specific sources to precipitation chemistry at a chosen receptor point;
- Estimation of transport of acidic-precipitation precursors across political borders;
- Estimation and prediction of air-quality modifications occurring as a consequence of the scavenging process;
- Site selection for precipitation-chemistry network sampling stations;
- Design of field studies of precipitation scavenging;

and

- Elucidation of mechanistic behavior of the scavenging process on the basis of field measurements.

In selecting an appropriate model, the user should review his intended application carefully with regard to the pollutant materials of interest, the time and distance scales, the processes in Figure C.3-2 covered, the source configuration, the precipitation type, and the mechanistic detail required. The question of pollutant materials is particularly important when precipitation acidity is of interest. Acidity in precipitation is determined by the presence of a multitude of chemical species, and in principle one must compute (via a model) the scavenging of each species and then estimate acidity on the basis of an ion balance:

$$[H^+] = \Sigma \text{ Anions} - (\Sigma \text{ cations other than } H^+). \quad \text{(C.3-1)}$$

Inorganic ions usually important in precipitation chemistry are itemized in Table C.3-4. Organic species play a secondary role in the acidification process, which appears to vary widely with region. Modeling of all of these species simultaneously requires substantial effort, and all "acid-precipitation" models up to the present have focused on only one or just a few of the more important species, with contributions of the others estimated on the basis of empiricism. Currently there is a tendency for newer models to accommodate larger numbers of these species; but complete modeling coverage will not be achieved in the foreseeable future.

Mechanistic detail is another important feature determining the basic composition of a scavenging model. A comprehensive mathematical description of the scavenging process can become rapidly overwhelming, and there is usually a need to represent these relationships in a comparatively simple, albeit approximate, manner. The process of consolidating complex behavior in this fashion is often referred to as lumping the system's parameters. The resulting simplified expressions are termed parameterizations. Consolidating the effects of nonmodeled species in empirical form, described in the preceding paragraph, is one example of lumping. Numerous other examples will arise throughout the remainder of this section.

This section will not attempt to provide the reader with a detailed treatise on how models should be

TABLE C.3-4 Some Inorganic Ions Important in Precipitation Chemistry[a]

Cations	Anions
H^+	
NH_4^+	Cl^-
Na^+	NO_3^-
K^+	$SO_3^=$
Ca^{++}	$SO_4^=$
Mg^{++}	$PO_4^=$
	$CO_3^=$

[a]All ions are presented here in their completely dissociated states. The reader should note, however, that various states of partial dissociation are possible as well (e.g., HSO_3^-, HCO_3^-).

formulated and applied.* The approach, rather, will be to develop a basic understanding of the fundamental elements of a scavenging model and then provide a systematic procedure for choosing and locating appropriate models from the literature. The following subsection discusses the basic conservation equations, which constitute the conceptual bases for scavenging models in general. This is followed in turn by two simple applications of these relationships, which are presented to illustrate usage and to define some terms commonly

*For the reader interested in more detailed pursuit of this area, the works by Hales (1983) and Slinn (1983) are recommended. The Hales reference is something of a beginner's primer, while Slinn's treatment delves substantially deeper into mechanistic detail. Together they constitute a reasonable starting point for understanding and modeling basic scavenging phenomena.

used in scavenging models. The final subsection attacks the problem of model selection, using a flow-chart approach, which is designed to guide the user to a valid choice in a systematic manner that avoids many of the pitfalls normally encountered on such endeavors.

3.4.2 Elements of a Scavenging Model

3.4.2.1 Material Balances

In Figure C.3-3 the various arrows between boxes correspond physically to streams of pollutant and/or water, and from this it is not difficult to realize that any characterization of this system must include material balances. Material balances thus form the underlying structure for all scavenging models. To formulate a material balance one simply visualizes some chosen volume of atmosphere and sums over all inputs and outputs of the substance in question.

Two basic types of material balance are possible:

1. "Microscopic" material balances, based on summation over a limiting small volume element of atmosphere;

and

2. "Macroscopic" material balances, based on summation over a larger volume element of atmosphere (e.g., a complete storm system).

Microscopic material balances invariably lead to differential equations, which must be integrated over finite limits to obtain practical results. Macroscopic balances result in mixed, integral, or algebraic equations. Again the choice of material-balance type depends on the specific modeling purpose at hand.

An important general form of the differential material balance for some chosen pollutant (denoted by subscript A) is given by the equations* (cf. Hales 1983)

Equations (C.3-2) and (C.3-3) are quite general in the sense that the velocity vectors denote velocity of pollutant (rather than that of the bulk media) and thus provide for all modes of transport (convective, diffusive, ...) without yet specifying how this transport is to occur. These equations are not yet time-smoothed; thus no closure assumptions have been applied at this point.

$$\frac{\partial c_{Ay}}{\partial t} = -\tilde{\nabla} \cdot c_{Ay}\tilde{v}_{Ay} - w_A + r_{Ay} \text{ (gas phase)} \qquad \text{(C.3.2)}$$

and

$$\frac{\partial c_{Ax}}{\partial t} = -\tilde{\nabla} \cdot c_{Ax}\tilde{v}_{Ax} + w_A + r_{Ax} \text{ (aqueous phase).} \qquad \text{(C.3.3)}$$

Here c_{Ay} and c_{Ax} denote concentrations of pollutant in the gaseous and condensed-water phases, respectively. The time rate of change of these concentrations within the differential volume element is related to the sum of inputs by (1) flow through the walls of the element, (2) interphase transport between the gaseous and condensed phases, and (3) chemical (and/or physical) reaction within the element. The $\tilde{v}$ terms in Equations (C.3-2) and (C.3-3) denote velocity vectors, while $\tilde{\nabla}\cdot$ is the standard vector divergence operator. The interphase transport term w_A accounts for all "attachment" processes (impaction, phoresis, diffusion, . . .) as well as any reverse phenomena such as pollutant-gas desorption, while the r terms denote chemical conversion rates in the usual sense. To formulate a usable model from these equations one needs to specify values for the functions v, w, and r and then solve differential equations (C.3-2) and (C.3-3) (subject to appropriate initial and boundary conditions) to obtain the desired concentration fields c_{Ay} and c_{Ax}. A simple example of this procedure is given in Section 3.4.2.

Energy Balances

Many terms in Equations (C.3-2) and (C.3-3), especially v_{Ax}, w_A, and r_{Ax}, depend strongly on the amount, state, and interconversion rates of condensed water; and it is important at this point to note that atmospheric water itself obeys material-balance expressions of this form. In selecting a scavenging model one often is confronted with the problem of deciding whether to estimate precipitation attributes and these related terms independently on the basis of assumptions or previous information or to attempt to compute the desired entities directly by solving appropriate forms of Equations (C.3-2) and (C.3-3).

If the latter of these alternatives is chosen, then the inclusion of an energy-balance equation is mandatory. This need arises because the evaporation-condensation process influences, and is influenced by, a variety of energy-related considerations. These include temperature influences on vapor pressure and latent-heat effects and can be incorporated in the model via an energy balance performed over the same element of atmosphere as that of the associated material balances. In microscopic form, a general expression of the energy balance (cf. Bird et al. 1960), is

$$\rho C_v \frac{\partial T}{\partial t} = - \nabla \cdot \tilde{h} - p \nabla \cdot \tilde{v} + \Gamma - D \ . \qquad \text{(C.3-4)}$$

Here the time rate of change of temperature is related to the sum of inputs by (1) flow through the walls of the element and (2) generation via (a) compression work, (b) latent heat effects, and (c) frictional dissipation. The vector terms h and v denote sensible heat flux and fluid velocity, respectively, while Γ and D pertain to latent heat and dissipation. ρ and C_v denote fluid density and specific heat in the usual sense. A straightforward example of the incorporation of Equation (C.3-4) for scavenging modeling purposes is given by Hales (1983).

Momentum Balances

Solutions to Equations (C.3-2)-(C.3-4) depend on the existence of some previous description of fluid velocity v [or v_{Ay} in the case of Equation (C.3-2)]. As was the case for the preceding parameters associated with the energy balance, velocity may be specified for the model on the basis of previous measurements or assumptions. Flow patterns in storm systems may be sufficiently complex to defy empirical specification, however, and the modeler may wish to compute the associated fields on the basis of a modeling approach. If this is to be done, a momentum-balance equation must be employed. In microscopic form the general momentum balance may be expressed (cf. Bird et al. 1960) as

$$\frac{\partial}{\partial t} \rho v = -\nabla \cdot \rho \tilde{v}\tilde{v} - \nabla p - \tilde{F}_v + \rho g. \qquad \text{(C.3-5)}$$

Here the time rate of change of momentum (ρv) is expressed as the sum of inputs by (1) flow through the walls of the element, (2) pressure forces, (3) viscous drag forces, and (4) gravitational forces. To apply Equation (C.3-5) for modeling purposes one specifies frictional, pressure, and gravitational terms and solves the differential equation subject to appropriate initial and boundary conditions to obtain fields of the velocity vector $\tilde{v}$. An example of application of Equation (C.3-5) for scavenging modeling purposes is given by Hane (1978).

Incorporation of energy and momentum balances Equations (C.3-4) and (C.3-5) into a scavenging model is a rather challenging exercise, and a relatively small number of models exist that apply these equations for this purpose. The usual tack is simply to "prespecify" the required parameters and proceed with material-balance calculations alone. Numerous examples of both types of models will be presented in Section 3.4.5.

3.4.3 Definitions of Scavenging Parameters

Four key parameters often arise in the context of scavenging models, and it is appropriate at this point to define these terms and indicate their general application. Reference to these entities as "parameters" is consistent with the usage applied in the previous section, in that they serve to "lump" the effects of a number of mechanistic processes in a simple formulation. These will be discussed sequentially in the following paragraphs.

The first parameter to be defined is the attachment efficiency. Also known as the capture efficiency, this term can be visualized most easily by considering a hydrometeor falling through a volume of polluted air space, as shown in Figure C.3-20. This hydrometeor sweeps out a volume of air during its passage; and attachment efficiency is defined as the amount of collected pollutant divided by the amount that was initially in this volume. The efficiency can exceed 1.0 if pollutant from outside the swept volume becomes attached to the drop.

From the discussion of attachment mechanisms in Section 3.2 it is seen that the attachment efficiency accounts for a multitude of processes. Usually the efficiency is less than 1; but mechanisms such as diffusion, electrical effects, and interception can give rise to larger values, especially when the collecting

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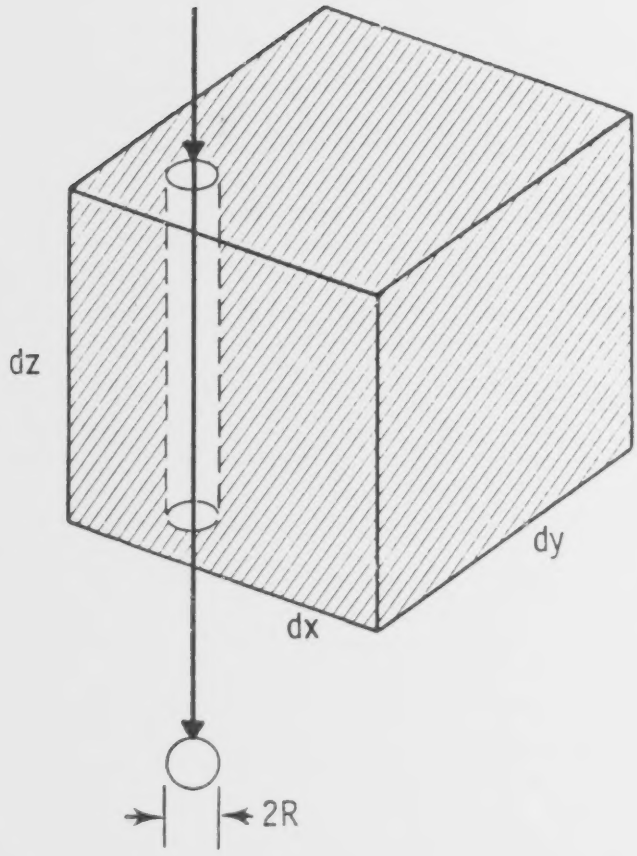


FIGURE C.3-20 Schematic of a scavenging Hydrometeor falling through a volume element.

element's fall velocity is small. Efficiencies can be negative if the element is releasing pollutant to the surrounding atmosphere, such as in the case of pollutant-gas desorption. Typical efficiencies for aerosol particles collected by raindrops are shown in Figure C.3.4.

Another important parameter is the scavenging coefficient. This entity is basically an expression of the law of mass action, and is defined by the form

$$\Lambda = \frac{w_A}{c_{AY}}, \qquad \text{(C.3-6)}$$

where [in a manner consistent with Equations (C.3-2) and C.3-3)] w_A is the rate of depletion of pollutant A from the gaseous phase by attachment to the aqueous phase in a differential volume element. This is similar to a rate expression for a first-order, irreversible chemical reaction, and as such it applies strictly only to irreversible attachment processes (e.g., aerosols or highly soluble gases). Λ can be related to the attachment efficiency E by the form (which assumes spherical hydrometeors)

$$\Lambda(a) = -\pi N_T \int_0^\infty R^2 v_z(R) E(R,a) f_R(R)\, dR, \qquad \text{(C.3-7)}$$

where a and R denote aerosol and hydrometeor radii, respectively, v_z is the hydrometeor fall velocity, and N_T and f_R are the total number and probability-density functions for the size-distributed hydrometeors residing in the volume element of Figure C.3-20 at any instant in time. From this one can note that Λ essentially extends the paramaterization over the total spectrum of hydrometeor sizes.

Atmospheric aerosol particles are typically distributed over extensive size ranges, and because of this it is often desirable to possess some sort of an effective scavenging coefficient, which represents a weighted average over the aerosol size spectrum. Figure C.3-21 presents a family of curves corresponding to such averages, which are based on assumed log-normal particle-size spectra, with different geometric standard deviations. From these curves one can observe that for the same geometric mean particle size, changes in <u>spread</u> of the size distribution can result in dramatic changes in the effective scavenging coefficient.

Inclusion of reversible attachment processes in a scavenging model usually involves utilization of the <u>mass-transfer coefficient</u>. This parameter can be defined in terms of the flux of pollutant moving from the scavenging element as

$$\text{Flux} = -\frac{K_y}{c}\left(c_{Ay} - h' c_A\right). \qquad \text{(C.3-8)}$$

Here K_y is the mass-transfer coefficient and c_A is the concentration, within the scavenging element, of

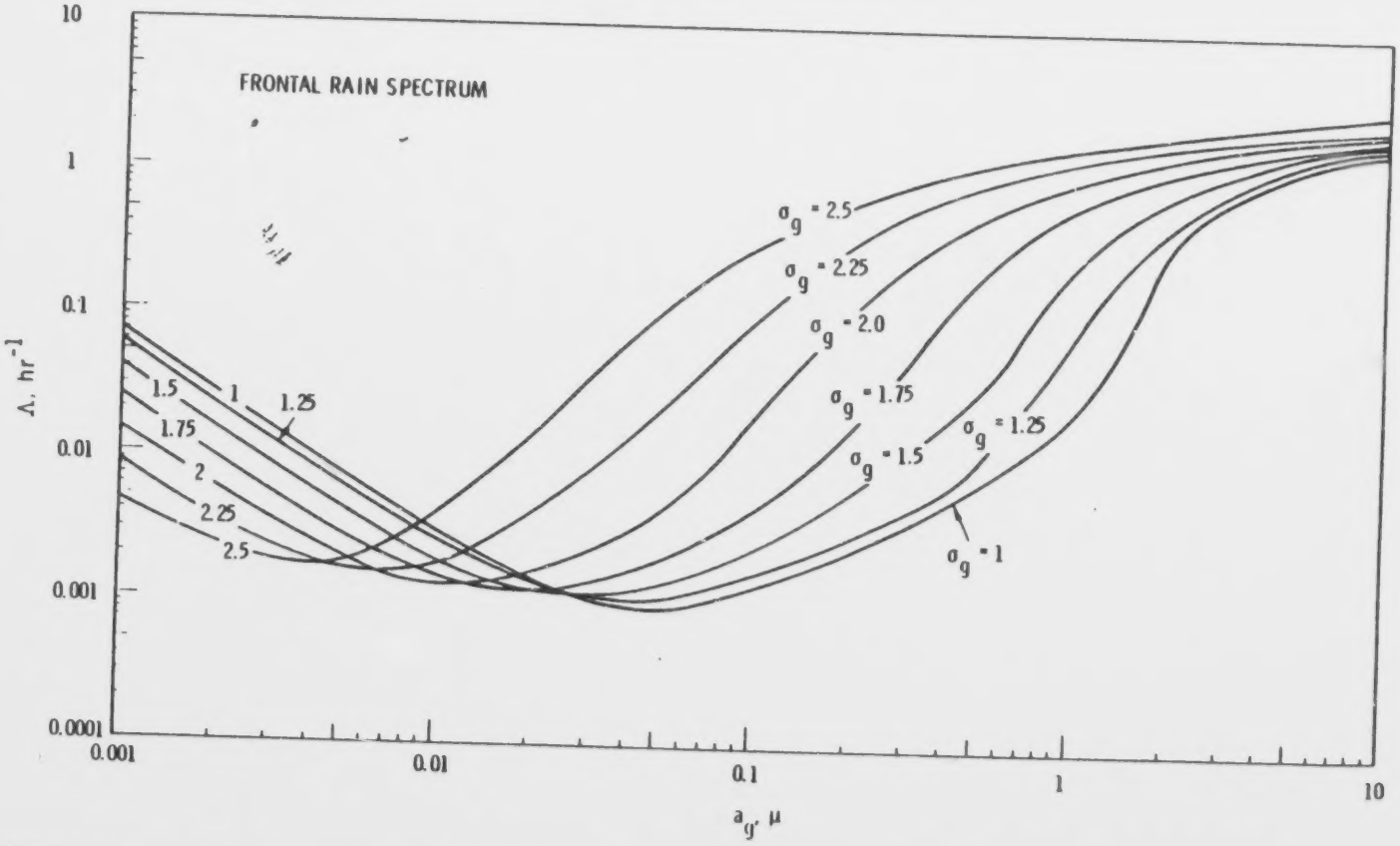


FIGURE C.3-21 Computed effective scavenging coefficients for size-distributed aerosols. Based on a log-normal aerosol radius distribution with geometric means and standard deviations a_g and σ_g. A typical frontal-rain drop-size spectrum is assumed. From Dana and Hales (1976).

collected pollutant. h' is essentially a solubility coefficient, which, when multiplied by c_A, produces a gas-phase equilibrium value. c is the molar concentration of air molecules, which appears in Equation (C.3-8) because of the manner in which K_y has been defined. Thus the flux can be either to the drop or away from it, depending on the relative magnitudes of the parenthetical terms. Equation (C.3-8) can be integrated over all drop sizes in a manner similar to that used in Equation (C.3-7) (cf. Hales 1972) to form the following expression for w_A:

$$w = \frac{4\pi N_T}{c} \int_0^\infty R^2 f_R(R) K_y(R) (c_{Ay} - h' \hat{c}_A) \, dR. \qquad (C.3\text{-}9)$$

The final scavenging parameter to be described here is the scavenging ratio. This entity is usually the result of a model calculation, rather than an input, and is defined by the form

$$\xi = \frac{\hat{c}_A}{c_{Ay}}, \qquad (C.3\text{-}10)$$

where C_A is the concentration of pollutant contained in a collected precipitation sample. ξ is a term that is immediately usable for a number of pragmatic purposes, because once its numerical value is known it can be applied directly to compute precipitation-chemistry concentrations on the basis of air-quality measurements. Tables of measured (Engelmann 1971) and model-predicted (Scott 1978) scavenging washout ratios have been published, although caution is advised in the application of these values. A simple example of scavenging-ratio application is given in the following section.

It is useful for the sake of visualization to discuss briefly the qualitative features of the scavenging parameters noted above. The parameter E is easy to visualize in the context of Figure C.3-20; it is, simply, the collection efficiency of an individual cloud or precipitation element and as such should be expected to fall numerically in the approximate range between zero and one. The scavenging coefficient Λ can be visualized as a first-order removal rate, in much the same manner as that of a first-order reaction-rate

coefficient. As such it may be utilized roughly as a characteristic time scale for wet removal. $\Lambda = 1$ h^{-1}, for example, would imply that the scavenging process will cleanse $100(1 - 1/e)$ percent of the pollutant in 1 h if conditions remain constant and competitive processes do not occur. From this one can note that 1 h^{-1} is a moderately large scavenging coefficient. Λs ranging from zero to 1 h^{-1} and beyond have been reported in the literature (cf. Figure C.3-21).

The mass-transfer coefficient K_y is essentially a normalized interfacial flux of pollutant between the atmosphere and an individual droplet. Little needs to be said here regarding magnitudes of K_y, except to note that a variety of different definitions of K_y exist, and one must be cognizant of these definitions when employing values obtained from outside sources. The washout ratio, ξ, is essentially a measure of the concentrating power of precipitation in its extraction of pollutant from the atmosphere. As will be noted in the next section, precipitation often has the ability to concentrate airborne pollution by a factor of a million or more. ξs ranging from below 100 up through 10^8 and higher have been reported in the literature.

The expected magnitudes and uncertainty levels associated with the scavenging parameters listed in this section depend strongly on the substance being scavenged and the environment in which the scavenging takes place. Large aerosol particles in below-cloud environments, for example, are characterized by scavenging efficiencies in the range of 1.0 (cf. Figure C.3-4), which can be estimated with relatively high precision. Smaller particles, especially those in the "Greenfield-gap" region, are much more difficult to simulate, and associated errors in estimated efficiencies may approach an order of magnitude or more. Errors in these efficiency estimates will of course be compounded by uncertainties in raindrop size spectra, if extended to scavenging coefficients via Equation (C.3-7). In the case of gases, the mass-transfer coefficient usually can be estimated to within a factor of 2 or less; again this error can be expected to compound when integrated over assumed raindrop size-spectra.

In the case of in-cloud scavenging of aerosols our capability for estimating transport parameters is seriously impeded, owing to the profusion of mechanisms and the complex environments involved. Typical

uncertainties in both Λ and ζ can be expected to approach an order of magnitude in some cases. Some appreciation for the factors influencing in-cloud scavenging coefficients can be obtained from the work of Slinn (1977), who attempts to evaluate theoretical, "storm-averaged" values for Λ. An idea of the magnitudes and uncertainties of ξ is given in Figure C.3-23.

In all cases involving reactive gases the values of E, Λ, and ξ are heavily contingent on the aqueous-phase chemical processes involved. Much remains to be accomplished in our understanding of aqueous-phase chemistry before a meaningful assessment of associated uncertainties is possible.

As a final note in this context it should be emphasized that uncertainties in scavenging parameters dictate uncertainties in scavenging calculations in a complex fashion and that errors associated with the microscopic phenomena can be either amplified or attenuated by their applications in macroscopic models to produce practical results. Uncertainties associated with macroscopic modeling applications will be discussed at some length in a later section.

3.4.4 Formulation of Scavenging Models: Simple Examples of Microscopic and Macroscopic Approaches

As noted previously, the description given in this document will refrain in general from deriving and applying scavenging models explicitly. This is too broad and complex a topic to be discussed in detail here, and the reader is referred to the previously cited literature for more detailed pursuit of this subject. For purposes of illustration, however, it is worthwhile to consider two simple examples of scavenging-model formulation, which demonstrate microscopic and macroscopic approaches to the problem. The present subsection addresses this task.

The microscopic material balance approach will be considered first. For this example it is useful to visualize an idealized situation where rain of known characteristics is falling through a stagnant volume of atmosphere, which contains a well-mixed, nonreactive pollutant with concentration c_{Ay}. The air velocity is known ($v = 0$) so solution of the momentum equation (C.3-5) is not required. The raindrop size distribution is presumed to remain constant; thus evaporation-

condensation and other energy-related effects are immaterial, and the energy equation (C.3-4) may be disregarded.

Since the pollutant is well mixed, no concentration gradients occur; thus the divergence term in Equation (C.3-2) is zero. Because of *nonreactivity the reaction* term is zero as well.

Now presume that the pollutant is an aerosol, whose attachment can be characterized in terms of the known scavenging coefficient Λ, using Equation (C.3-6). The corresponding reduced form of Equation (C.3-2) is, then,

$$\frac{\partial c_{Ay}}{\partial t} = -\Lambda c_{Ay}. \qquad \text{(C.3-2a)}$$

Given some initial pollutant concentration c_{Ayo}, Equation (C.3-2a) can be integrated to obtain the form

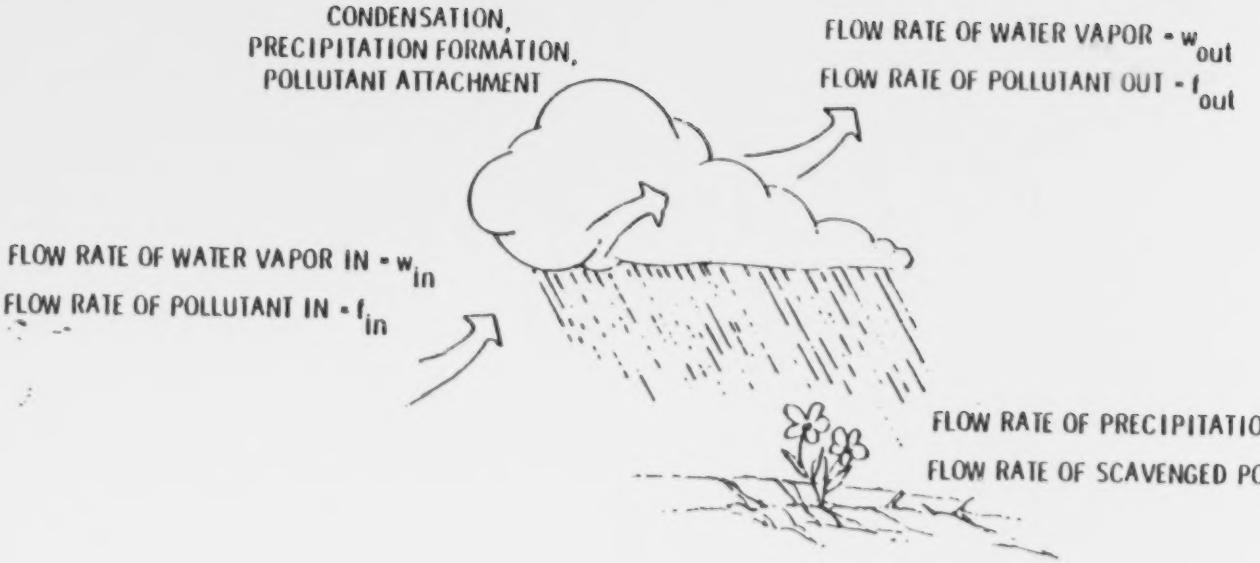
$$c_{Ay}(t) = c_{Ayo} \exp(-\Lambda t), \qquad \text{(C.3-11)}$$

which expresses the decrease of the gas-phase pollutant concentration with time. Counterpart expressions for rainborne concentrations may be derived by subjecting Equation (C.3-3) to a similar treatment.

The reader is cautioned to consider this treatment as an example only and to recognize that actual atmospheric conditions seldom conform to the idealizations invoked above. Gas-phase concentrations are usually not uniformly distributed in space, raindrop characteristics are usually not invariant with time, wind fields are usually not well characterized by $v = 0$. Λ is usually not a time-independent constant, and many pollutants are usually not well characterized by the washout coefficient approximation, anyway. The pollutant often is not unreactive. Examples of existing models where these constraints are relaxed in various ways are presented in the following subsection.

Figure C.3-22 illustrates the formulation of a macroscopic type of scavenging model. Here, in contrast to the differential-element approach, the material balances are formulated around a large volume element, in this case a total storm. If one denotes concentrations and flow rates of water and pollutant as follows:

c_{Ay} = airborne concentration of pollutant,
H = airborne concentration of water vapor into cloud,


CONDENSATION,
PRECIPITATION FORMATION,
POLLUTANT ATTACHMENT
FLOW RATE OF WATER VAPOR - w_{out}
FLOW RATE OF POLLUTANT OUT - f_{out}
FLOW RATE OF WATER VAPOR IN - w_{in}
FLOW RATE OF POLLUTANT IN - f_{in}
FLOW RATE OF PRECIPITATION OUT - W
FLOW RATE OF SCAVENGED POLLUTANT OUT - F

DEFINITIONS OF EFFICIENCIES:

WATER REMOVAL

$$\epsilon_p = \frac{W}{w_{in}}$$

POLLUTANT REMOVAL

$$\epsilon = \frac{F}{f_{in}}$$

FIGURE C.3-22 Schematic of a typical macroscopic material balance.

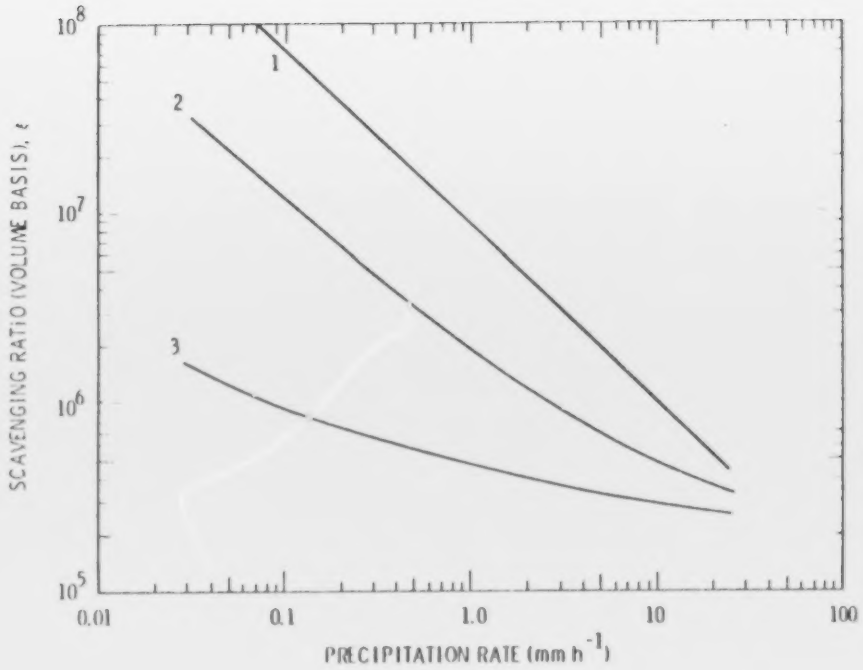


FIGURE C.3-23 Scott's scavenging ratio curves: 1, convective storms; 2, warm, non-convective storms; 3, cold storms, where Bergeron-Findeisen process is active (Scott 1978).

$\hat{C}_A$ = concentration of scavenged pollutant in rainwater,
ρ_w = density of condensed water,
w_{in} = flow rate of water vapor into the storm,
w_{out} = flow rate of water vapor out of the storm,
f_{in} = flow rate of pollutant into the storm,
f_{out} = flow rate of pollutant out of the storm,
W = low rate of precipitation out of the storm,
F = flow rate of scavenged pollutant out of the storm,

then extraction efficiencies for water vapor and pollutant can be defined, respectively, as

$$\varepsilon_p = \frac{W}{w_{in}} \qquad \text{(C. 3-11)}$$

and

$$\varepsilon = \frac{F}{f_{in}}. \qquad \text{(C.3-12)}$$

If one further performs material balances over this storm system for pollutant and water vapor, and then combines the two, the following form is obtained:

$$\xi = \frac{\hat{C}_A}{C_{AY}} = \frac{\varepsilon_p \rho_w}{\varepsilon H}, \qquad \text{(C.3-13)}$$

where the scavenging ratio, ξ, was defined earlier in Section 3.4.3.

Equation (C.3-13) is an important result in the sense that it demonstrates once again the strong linkage between water-extraction and pollutant-scavenging processes. If both occur with equal efficiency ($\varepsilon_p = \varepsilon$),* for example, then

$$\xi = \frac{\rho_w}{H} \simeq 10^5\text{-}10^6 \qquad \text{(C.3-14)}$$

Experimentally measured scavenging ratios often fall in this range, although wide variability often may be observed.

Utilizing a rather involved series of arguments pertaining to cloud-physics processes and attachment mechanisms, Scott (1978) has created a family of curves expressing aerosol scavenging ratio as a function of precipitation rate. Shown in Figure C.3-23, curves 1, 2,

*There is no direct reason to expect that ε_p should be similar to ε in magnitude. In the absurd circumstance where all the pollutant were concentrated into one particle, for example, then scavenging of that pollutant by a very light rainfall would yield $\varepsilon \simeq 1.0 >> \varepsilon_p$. Conversely, a large storm processing an insoluble gaseous pollutant (SF_6, say) would provide $\varepsilon \simeq 0 << \varepsilon_p$. For practical conditions involving acid-forming aerosols, however, the scavenging of water vapor and pollutant appear to be sufficiently related to allow $\varepsilon_p \simeq \varepsilon$ to be employed as an approximate rule of thumb.

and 3 pertain, respectively, to convective storms, nonconvective warm-rain process storms, and cold storms where the Bergeron-Findeisen process is active.

A major assumption of Scott's analysis is that the pollutant is ingested by the storm in the form of aerosol particles that are active as cloud condensation nuclei. The analysis also assumes a steady-state storm system and complete vertical mixing of pollutant between the storm height and the surface. Under such conditions Scott's curves can be considered as reasonably good estimators of actual scavenging behavior. More elaborate systems, involving reactive pollutants, gases, and nonhomogeneous systems are discussed in references given in the following section.

3.4.5 Systematic Selection of Scavenging Models: A Flow-Chart Approach

Hales (1983) has suggested a flow-chart approach to aid in the process of scavenging-model selection. Presented as a decision tree in Figure C.3-24, the user proceeds by answering a series of questions that relate to the model's intended use, the temporal and geographical scales, the pollutant characteristics, the choice between macroscopic and microscopic material balances, and the type of conservation (i.e., material, energy, momentum) equations involved. Various pathways through this decision tree are discussed in the original reference.

Proceeding through Figure C.3-24 in this manner the user can arrive at simple or complex end points, depending on the nature of his particular application. A trivial example is pathway 1-5-6, which instructs the user to disregard modeling totally and rely solely on past measurements. The simple microscopic-balance example of Section 3.4.4 can be traced through pathway 1-2-7-8-21-23-15-16.

Table C.3-5 presents an itemization of some currently available models, which can be related directly to the pathways of Figure C.3-24. This provides the reader with a rapid and efficient means of access to current modeling literature, while minimizing the chance of pitfall encounters that can arise from the inadvertent invocation of inappropriate physical constraints. For a more definitive description of this model selection process, the reader is referred to Hales's original reference.

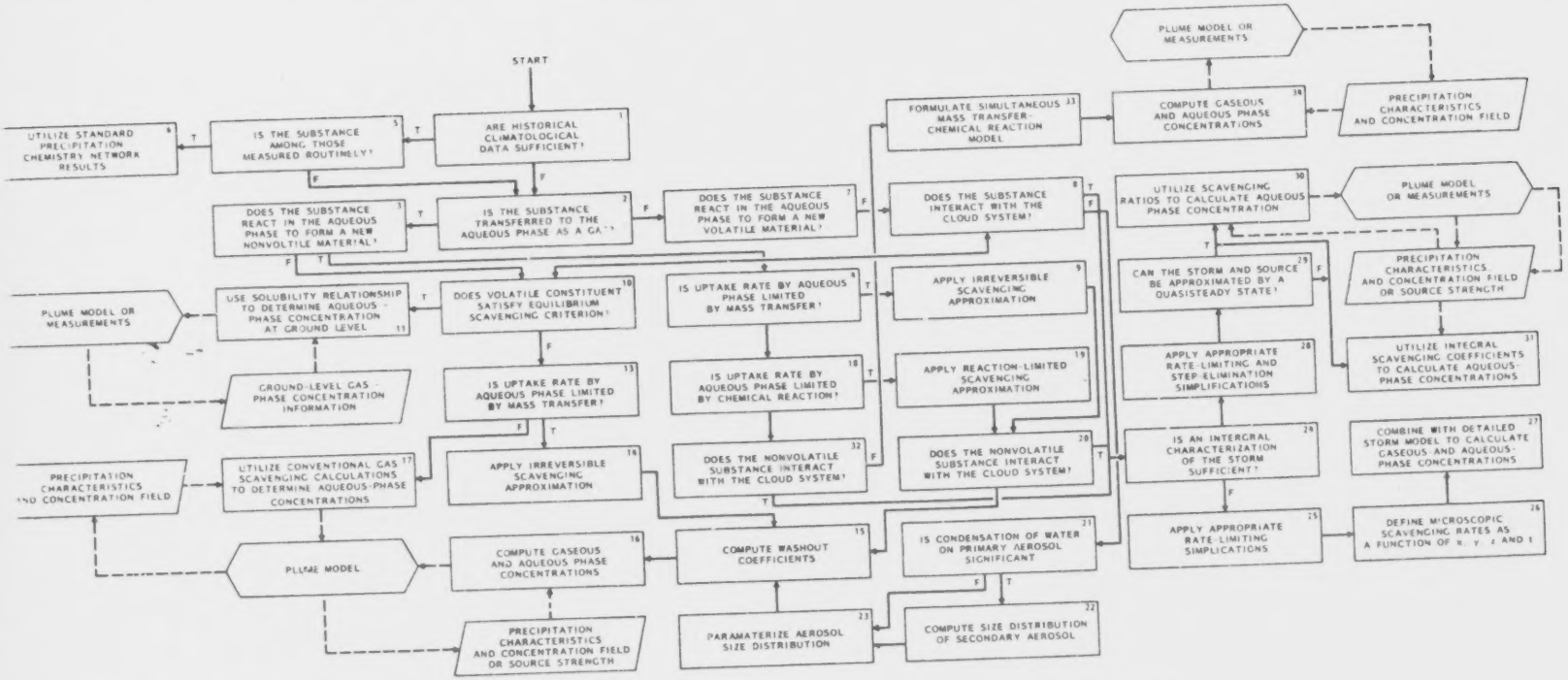

START
ARE HISTORICAL CLIMATOLOGICAL DATA SUFFICIENT?
IS THE SUBSTANCE AMONG THOSE MEASURED ROUTINELY?
UTILIZE STANDARD PRECIPITATION CHEMISTRY NETWORK RESULTS
IS THE SUBSTANCE TRANSFERRED TO THE AQUEOUS PHASE AS A GAS?
DOES THE SUBSTANCE REACT IN THE AQUEOUS PHASE TO FORM A NEW NONVOLTILE MATERIAL?
DOES THE SUBSTANCE REACT IN THE AQUEOUS PHASE TO FORM A NEW VOLATILE MATERIAL?
DOES THE SUBSTANCE INTERACT WITH THE CLOUD SYSTEM?
FORMULATE SIMULTANEOUS MASS TRANSFER-CHEMICAL REACTION MODEL
COMPUTE GASEOUS AND AQUEOUS PHASE CONCENTRATIONS
PLUME MODEL OR MEASUREMENTS
PRECIPITATION CHARACTERISTICS AND CONCENTRATION FIELD
UTILIZE SCAVENGING RATIOS TO CALCULATE AQUEOUS PHASE CONCENTRATION
PLUME MODEL OR MEASUREMENTS
CAN THE STORM AND SOURCE BE APPROXIMATED BY A QUASISTEADY STATE?
PRECIPITATION CHARACTERISTICS AND CONCENTRATION FIELD OR SOURCE STRENGTH
APPLY APPROPRIATE RATE-LIMITING AND STEP-ELIMINATION SIMPLIFICATIONS
UTILIZE INTEGRAL SCAVENGING COEFFICIENTS TO CALCULATE AQUEOUS-PHASE CONCENTRATIONS
IS AN INTEGRAL CHARACTERIZATION OF THE STORM SUFFICIENT?
COMBINE WITH DETAILED STORM MODEL TO CALCULATE GASEOUS AND AQUEOUS-PHASE CONCENTRATIONS
APPLY APPROPRIATE RATE-LIMITING SIMPLICATIONS
DEFINE MICROSCOPIC SCAVENGING RATES AS A FUNCTION OF x, y, z AND t
IS UPTAKE RATE BY AQUEOUS PHASE LIMITED BY MASS TRANSFER?
APPLY IRREVERSIBLE SCAVENGING APPROXIMATION
IS UPTAKE RATE BY AQUEOUS PHASE LIMITED BY CHEMICAL REACTION?
APPLY REACTION-LIMITED SCAVENGING APPROXIMATION
DOES THE NONVOLATILE SUBSTANCE INTERACT WITH THE CLOUD SYSTEM?
DOES THE NONVOLATILE SUBSTANCE INTERACT WITH THE CLOUD SYSTEM?
IS CONDENSATION OF WATER ON PRIMARY AEROSOL SIGNIFICANT
COMPUTE SIZE DISTRIBUTION OF SECONDARY AEROSOL
PARAMATERIZE AEROSOL SIZE DISTRIBUTION
COMPUTE WASHOUT COEFFICIENTS
COMPUTE GASEOUS AND AQUEOUS PHASE CONCENTRATIONS
PRECIPITATION CHARACTERISTICS AND CONCENTRATION FIELD OR SOURCE STRENGTH
PLUME MODEL
UTILIZE CONVENTIONAL GAS SCAVENGING CALCULATIONS TO DETERMINE AQUEOUS-PHASE CONCENTRATIONS
PRECIPITATION CHARACTERISTICS AND CONCENTRATION FIELD
IS UPTAKE RATE BY AQUEOUS PHASE LIMITED BY MASS TRANSFER?
APPLY IRREVERSIBLE SCAVENGING APPROXIMATION
USE SOLUBILITY RELATIONSHIP TO DETERMINE AQUEOUS-PHASE CONCENTRATION AT GROUND LEVEL
DOES VOLATILE CONSTITUENT SATISFY EQUILIBRIUM SCAVENGING CRITERION?
GROUND-LEVEL GAS-PHASE CONCENTRATION INFORMATION
PLUME MODEL OR MEASUREMENTS

FIGURE C.3-24 Flow chart for scavenging calculations.

3.5 PRACTICAL ASPECTS OF SCAVENGING MODELS: UNCERTAINTY LEVELS AND SOURCES OF ERROR

Quantitative assessment of the predictive capability of present wet-removal models is a complex task and is well beyond the scope of this document. There are, however, a number of general statements that are highly useful in focusing in on this question and in providing insights pertaining to model reliability. These are itemized sequentially below.

- The predictive capability of a scavenging model is strongly contingent on its desired application.

As noted in Section 3.4.1, there exists a variety of different applications of scavenging models, and some are much more difficult to fulfill than others. One can, for example, employ existing regional models to reproduce distributions of annually averaged, wet-deposited, sulfate ion in eastern North America with moderate success. If one is charged with the task of relating specific sources to deposition at a chosen receptor site, however, our predictive capability can be expected to be relatively imprecise. Similarly, if one is expected to forecast the change in deposition that would occur in response to some future change in emissions, then the associated uncertainty level would be very high indeed. The question of nonlinear response is of paramount importance in this last application.

A large component of our uncertainty in predicting source attribution and transient response is based simply on the fact that we do not have adequate data bases for testing model performance for these applications. Our present models may in actuality be better predictors in this respect than anticipated; but because we have no immediate way of confirming this our uncertainty level remains high.

Regardless of the above considerations it should be emphasized strongly that the first step in scavenging model evaluation must be the precise definition of the intended uses of the model. All subsequent efforts will be confounded in the absence of this focal point.

- The predictive capability of a scavenging model is dependent on the choice of model.

At first sight this appears to be a self-evident and trivial statement. A profusion of scavenging models exists, however, and it is not at all difficult to choose

TABLE C.3-5 Pertinent Literature References for Wet-Removal Models

Model	Type of Balance Equations	Mechanisms
1. Classical washout coefficient	Material (differential)	Irreversible attachment
2. Distributed washout coefficient	Material (differential)	Irreversible attachment
3. "Two-stage" nucleation-accretion	Material (differential)	Irreversible attachment
4. Nonreactive gas scavenging	Material (differential)	Reversible attachment
5. Reactive gas scavenging	Material (differential)	Reversible attachment with aqueous-phase reaction
6. In-cloud aerosol scavenging	Material (differential)	Irreversible attachment
7. In-cloud aerosol scavenging	Material (integral)	Irreversible or reversible attachment
8. In-cloud reactive gas and aerosol scavenging	Material (differential)	Transport, reaction, and deposition
9. In-cloud reactive gas and aerosol scavenging	Material (integral)	Irreversible or reversible attachment with chemical reaction
10. Composite analytical	Material (differential)	Transport, reaction, and deposition
11. Composite trajectory	Material (differential)	Transport, reaction, and deposition
12. Composite grid	Material (differential)	Transport, reaction, and deposition
13. Composite statistical	Material	Transport, reaction, and deposition
14. Nonreactive	Material energy and momentum (differential)	Irreversible attachment, nonreactive
15. Reactive	Material and energy (differential)	All modes of scavenging including chemical reaction

Typical Application	Pertinent References
Below-cloud scavenging of aerosols and reactive gases	Chamberlain (1953), Engelmann (1968), Fisher (1975), Scriven and Fisher (1975), Wangen and Williams (1978)
Below-cloud scavenging of size-distributed aerosols	Dana and Hales (1976), Slinn (1982)
Condensation-enhanced below-cloud scavenging of aerosols	Radke et al. (1978), Slinn (1982)
Below-cloud scavenging of nonreactive gases	Barrie (1978), Hales et al. (1973), Slinn (1974b)
Below-cloud scavenging of reactive gases	Adamowitz (1979), Drewes and Hales (1982), Durham et al. (1981), Hill and Adamowitz (1977), Overton et al. (1979)
Scavenging in storm systems (nonreactive)	Dingle and Lee (1973), Junge (1963), Klett (1977), Lange and Knox (1977), Slinn (1982), Storebo and Dingle (1974)
Scavenging in storm systems	Engelmann (1971), Gatz (1972), Hales and Dana (1979), Scott (1978), Slinn (1982)
Scoping studies	Gravenhorst et al. (1975), Omstedt and Rodhe (1978)
Interpretation of study data	Scott (1982)
Regional-scale deposition	Astarita et al. (1979), Fay and Rosenzweig (1980)
Regional-scale deposition	Bass (1980), Bhumralker et al. (1980), Bolin and Persson (1975), Eliassen (1978), Fisher (1978), Hales (1977), Heffter (1980), Henmi (1980), Kleinman et al. (1980), McNaughton et al. (1981), Patterson et al. (1981), Sampson (1980), Shannon (1981), Voldner (1982)
Regional-scale deposition	Carmichael and Peters (1981), Lamb (1981), Lavery et al. (1980), Lee (1981), Liu and Durran (1977), Prahm and Christensen (1977), Wilkening and Ragland (1978)
Scoping studies and life-time assessment	Rodhe and Grandell (1972)
In-cloud scavenging analysis	Hane (1978), Kreitzburg and Leach (1978), Molenkamp (1974)
In-cloud scavenging analysis	Hales (1982a)

an inappropriate candidate inadvertently. Such inappropriate selections have on occasion resulted in reported calculations that have been in error by several orders of magnitude.

This component of error may of course be totally eliminated by selection of the most appropriate model for the intended application. The flow chart presented in Figure C.3-24 is a useful guide for this purpose, especially for those only casually familiar with the field.

• The predictive capability of a scavenging model depends strongly on the processes modeled.

As noted in the context of Figure C.3-2 a scavenging model may encompass one, several, or all of the steps in the composite wet-removal sequence. If only a small portion of this sequence is being considered, the model depends heavily on information supplied from the remaining components. This information may originate from assumptions, from empirical measurements, or from the output of other models.

Assuming that all input information is error-free, then it may be stated generally that, the more steps in Figure C.3-2 encompassed by a given model, the greater will be its predictive uncertainty. This is simply a consequence of propagating errors and must be considered as a primary factor when addressing the validation of wet-removal calculations.

• The predictive capability of a scavenging model is dependent on its areal range.

This statement is largely a corollary of the one immediately above. As a scavenging model is extended to, say, a regional scale it is forced to include essentially all the components of Figure C.3-2. As noted previously, this is likely to increase uncertainty levels appreciably.

• The predictive capability of a scavenging model is contingent on its temporal averaging time.

Owing to the propensity of stochastic phenomena to average out to mean values, the predictive capabilities of (especially regional) scavenging models can be expected to improve somewhat as averaging times increase. This improvement is, of course, gained at the expense of sacrificing temporal resolution, and a value judgment is necessary (again requiring a precise

definition of intended model application) at this juncture.*

This observation should be tempered by the fact that, in addition to random errors, scavenging models can be expected to possess substantial systematic biases. In general these biases do not decrease with averaging time and in fact many lead to cumulative discrepancies on occasion. Examples of systematic errors are biases in trajectory calculations and artificial offsets induced by the superimposition of random events on nonlinear processes. Again the seriousness of such factors is heavily contingent on the intended model application.

In general summary it may be stated that several important factors lead to widely varying levels of uncertainty in scavenging-model predictions. One may predict, for example, the scavenging of SO_2 from a local power-plant plume using existing models and expect to match measured results within a factor of 2. On the other hand, similar predictions of, say, the fraction of sulfate at a given receptor that originated from some particular source can be expected to have orders-of-magnitude associated uncertainty. Both a comprehensive model-evaluation effort and a substantially improved data base will be required before this situation can be remedied to any appreciable extent.

3.5 CONCLUSIONS TO SECTION 3

This section has provided an overview of meteorological processes contributing to wet removal of pollutants and has summarized the current state of our capability to describe these complex phenomena in mathematical form. Because of the magnitude of this problem it has been necessary to refrain from detailed descriptions of models and modeling techniques; rather, we have chosen to describe the general mathematical basis for wet-removal modeling, to give two simple examples of direct application, and then to supply the reader with a means

*This issue is especially pertinent in view of the contention, often voiced by some scientists within the acid-precipitation effects community, that temporally averaged results (averaging times of a few months or more) are totally adequate for assessment purposes.

for efficiently pursuing the available literature for specific applications of interest.

In conclusion to this discussion it is appropriate to summarize the state of these calculational techniques by asking the following questions:

- Just how accurate and valid are current wet-removal modeling techniques as predictions of precipitation chemistry and wet deposition; that is, how well do they fulfill the needs itemized in Section 3.4.1?
- What must be accomplished before the present capabilities can be improved?

The answers to these questions are somewhat mixed. Certainly the techniques discussed in this section, if used appropriately, are capable of order-of-magnitude determinations in many circumstances; and under restricted conditions they can even generate predictions having factor-of-2 accuracy or better. Moreover, there is ample explanation in existing theories of wet removal to account easily for the spatial and temporal variabilities observed in nature.

These capabilities, however, cannot be considered to be satisfactory in the context of current needs. The noted ability to explain spatial and temporal variability on a semiquantitative basis has not resulted in a large competence in predicting such variability in specific instances. Moreover, we possess little competence in identifying specific sources responsible for wet deposition at a given receptor site. Finally, the order-of-magnitude predictive capability noted above can hardly be judged satisfactory for most assessment purposes. In reviewing the discussions of this section against the backdrop of these deficits, several research needs become apparent. The most important of these are itemized in the following paragraphs.

- Much more definitive information is needed with regard to the scavenging efficiencies of submicrometer aerosols, for both rain and snow. Especially important in this regard is the effect of condensational growth of such aerosols in below-cloud environments.
- We need to know much more about aqueous-phase conversion processes, which are potentially important as alternate mechanisms resulting in the presence of species such as sulfate and nitrate in precipitation. Since virtually nothing is known at present regarding the

chemical formation of such species in clouds and precipitation, there is a tendency to lump these effects with physical removal processes in most modeling efforts, expressing them in terms of pseudo scavenging coefficients or collection efficiencies. Such phenomena must be resolved in finer mechanistic detail than this before a satisfactory treatment is possible, and this requires a knowledge of chemical transformation processes that is much more advanced than existing at present.

- Much more extensive understanding of the competitive nucleation capability of aerosols in in-cloud environments is needed, especially for those substances that do not compete particularly well in the nucleation process. The influence of aerosol-particle composition--especially for "internally mixed aerosols"*--is particularly important in this regard.
- The identification of specific sources responsible for chemical deposition at a given receptor location requires that we possess a much more accomplished capability to describe long-range pollution transport. Progress in this area during recent years has been encouraging, but much more remains to be achieved before we have a proficiency that is really satisfactory for reliable source-receptor analysis.
- We still need to enhance our understanding of the detailed microphysical and dynamical processes that occur in storm systems. Besides providing required knowledge of basic physical phenomena, such research is important in providing valid parameterizations of wet removal for subsequent use in composite regional models.

As a final note, it is useful to reflect once again on the fact that scavenging modeling research--as treated in this section--has been in a rather continuous state of development over the past 30 years. While progress has been indeed significant during this period, a number of important and unsolved problems still exist. Accordingly, one must use this perspective in assessing our rate of advancement during future years. Reasonable progress in resolving the above items can be expected over the next decade; but the complexity of these problems demands that a serious and sustained effort be applied for this purpose.

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*Those containing individual particles composed of a mixture of chemical species.

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APPENDIX C: COMMENTS OF JACK G. CALVERT AND ALAN LAZRUS ON THE INFORMAL "REVIEW" OF THE NRC COMMITTEE REPORT ON ACID DEPOSITION SUBMITTED BY THE BERNARD MANOWITZ GROUP

Recently we received a copy of a "review" of our NRC deposition report which was prepared under the guidance of Bernard Manowitz (Brookhaven National Laboratory) by selected representatives of three national laboratories supported by the Department of Energy: Brookhaven, Battelle Pacific Northwest, and Argonne. These "reviews" were sent to the National Academy of Sciences, to me, to various members of the Interagency Task Force on Acid Rain and others; my friends in industry and government tell me that the report was widely circulated to the leadership and research arms of the power industry and related industrial operations throughout the United States and within the extensive communications pipeline of the administrative and legislative staffs of the Federal and State governments. The original report has subsequently been sanitized and presented to Representatives Fuqua, Scheuer, and Lloyd with additional comments from a group at Oak Ridge National Laboratory; this has been or will be introduced into the Congressional Record. We did not have access to the revised version of the "review" of the Manowitz group until a few days ago, so we will necessarily respond to the original "review" which has received wide circulation. The scientific questions remain unchanged in the latest version. I have submitted as Appendix D of the formal report for the Congressional Record, the original Manowitz group "review" which should be published as part of the record of this hearing. The comments given here represent those of only two of our NRC acid deposition committee, myself (Jack G. Calvert) and Alan Lazrus, since time did not allow the participation of the entire committee.

A Congressional hearing or the Congressional Record is not well suited to scientific debate. In the long run scientific issues cannot be influenced by

legal or legislative actions or by pressures from business, conservation groups, or other special interest groups, including that headed by Manowitz. The scientific world does not accept blindly the stated opinions of any scientist or group of scientists, including the NRC committee on acid deposition, without extensive testing and checking of the ideas proposed. Fortunately for science the unresolved issues will be decided after extensive research and suitable discussions in the refereed scientific literature. However, I feel that the Manowitz group and its informal "review" cannot be disregarded by me since their "review" has been presented to a wide and important audience as a "scientific" endeavor which disputes some key aspects of the NRC acid rain report. Although it is difficult to sort out the science from the rhetoric in some of these "reviews", we will focus on what we judge to be the important scientific questions which have been raised.

First, the use of the Hubbard Brook data by the NRC acid deposition committee and our conclusions derived from them have been questioned. A reviewer designated as "D" has attempted to treat these and other data not available to the general scientific community, using a multiple linear regression analysis of "adjusted" data. He concluded that there is no 1:1 correspondence between sulfur oxide emissions and sulfate deposition at Hubbard Brook. However his conclusions must be rejected since his methods of data treatment and his arguments are flawed scientifically. The detailed analysis of the errors in his treatment have been submitted as part of the record of this hearing along with the original Manowitz group "review". It suffices for present purposes to indicate that he has made two serious errors. He made an inappropriate choice of variables in the multiple linear regression analysis, and he "corrects" the observed data using a long-range model and assumptions about the source regions based upon "local" winds (measured 100 miles from Hubbard Brook on the coast of Maine) which are definitely incorrect.

Given the lack of reliable models to predict accurately the source regions for the receptor site of Hubbard Brook, it is not scientifically sound to attempt the treatment of emissions and sulfate concentration at remote sites given by reviewer "D".

The conclusion of the NRC committee, namely that a large decrease in sulfate deposition at Hubbard Brook corresponded in time with a large decrease in sulfur dioxide emissions in the Northeast, is as definitive a statement as was permitted by the data. Certainly the attempt to be more quantitative made by reviewer "D" is seriously flawed scientifically, and must be rejected. His treatment of the sulfate to nitrate ratios which appears in the revised version of the Manowitz group "review" suffers from many of the same problems, and the results are of uncertain quality.

Reviewer "E" questioned the near constancy of the $SO_4^{=}/NO_3^{-}$ ratios observed over wide areas of eastern North America in the annual averaged deposition data. His comments show such an extraordinary degree of rancorous disdain and distortion that one should not honor them with a response. However our analysis of his comments (placed in the written material for the Congressional Record) shows no scientific basis for his claim. Instead a serious distortion of the observations is evident.

Our conclusion that reliability for present long-range transport and transformation models is low in relating specific sources with receptor sites has been questioned by one of the Manowitz "review" group ("F"), yet apparently it is supported by others of this group. The objection is surprising since our view seems to be the consensus view of the NRC committee on acid deposition, the Interagency Task Force on Acid Rain (of which the Manowitz group is some undefined part), and the Acid Rain Peer Review Panel.

c) The temperatures employed by reviewer D are monthly averages collected at a location 75 miles from Hubbard Brook. Data from weekly precipitation samples were regressed against monthly averaged temperatures, even though there is no model based on physical and chemical principles which is capable of realistically relating surface temperatures to the incorporation of sulfate in rain.

The correlation coefficient is a measure of how much variance in sulfate concentrations is attributable to the variance of sulfur dioxide emissions. Reviewer D's treatment of the data is an attempted mathematical reduction of the variance of the data, with the likely intent of removing portions of the variance not dependent on sulfur oxide emissions. However the parameters selected to accomplish this goal are themselves not physically unrelated to sulfur oxide emissions or to the transport of sulfur oxide emissions to remote sites. The data adjustments applied by reviewer D are on the one hand likely to remove some variance related to sulfur dioxide since the parameters used are related to sulfur dioxide emissions. On the other hand if these are inappropriate variables for the adjustment, then they will lead to nothing but an increase in the error component in the adjusted sulfate record. One must question the effectiveness of the adjustment when the mean for the adjusted record is 42% greater than that actually observed at Hubbard Brook. Note that including reviewer D's estimates of annual averages of sulfur dioxide emissions does not alter the result which we reported; these correlate with the observed sulfate concentration at the 95% confidence level. After "adjustment" of the actual data, there is no longer a correlation. Of course we did not have access to the emissions data estimates for each year so there was no way we could use more than the limited data made available to us by the Interagency Task Force on Acid Rain.

In our view there is a second major error in reviewer D's statistical treatment. In order to "correct" the observed data he models the long-range

transport of sulfur dioxide in a most unrealistic way. Based upon the failure to detect a strong correlation between local wind direction and sulfate observed in precipitation at Hubbard Brook, reviewer D surmised that sources from all directions can effect Hubbard Brook depositions equally and that their impacts are functions of the source strength and source distance alone. This is not an acceptable hypothesis. Furthermore reviewer D has chosen to use as representative of Hubbard Brook the local wind directions from a distant station on the coast of Maine, about 100 miles from Hubbard Brook, New Hampshire. A great many studies have shown the error in the use of even the actual local wind directions as an indicator of source region for long-range transport. In fact, the calculation of back trajectories of air parcels which bring precipitation to Hubbard Brook, suggest that less than 10% of the air masses come from the Midwest or EPA region V (Munn, et al., 1982). Nevertheless, since variations in the emission rates for sulfur dioxide in region V are much greater than the variations in the emission rates for regions I or II (New England), and the distance from Hubbard Brook to the centroid of emissions in Region V is several-times greater than that of regions I or II, the contribution of the Midwest in reviewer D's model is comparable to that of the Northeast. This is totally inconsistent with the actual air parcel trajectory analyses. Thus the lack of statistically significant correlation between sulfur dioxide "adjusted" values and any other variable cannot be justified scientifically or interpreted as physically realistic. Both the NRC acid deposition committee report and the full report of the Interagency Task Force on Acid Rain state that no present model describes in a quantitative fashion the relative impact of sulfur dioxide sources as a function of their strength and distance. Given the lack of such reliable models, it is not scientifically sound to attempt the treatment of emissions and sulfate concentration at remote sites given by reviewer D.

The conclusion of the NRC committee, namely that a large decrease in sulfate deposition at Hubbard Brook corresponded in time with a large decrease in sulfur

dioxide emissions in the Northeast, is as a definitive a statement as was permitted by the data. Certainly the attempt to be more quantitative made by reviewer D is seriously flawed scientifically, and it must be rejected. His treatment of sulfate to nitrate ratios which appears in the revised version suffers from the same problems, and the results are of uncertain quality.

Reviewer D is critical because of the inexact correspondence between the molar ratios of SO_2/NO_x in emissions and $SO_4^=/NO_3^-$ in wet deposition. However the report recognizes the discrepancies and deals with them in the following excerpt from this report:

> It is surprising to note in addition that the annually averaged molar ratios in precipitation at the MAP3S, NADP, and CANSAP sites in eastern North America resemble the molar ratios of SO_2 to NO_x emissions averaged on an annual basis by state or province (Figure 4.18). The emission ratios are somewhat higher than the deposition ratios in the Ohio and Tennessee River Valleys. Similarly, the emission ratios are somewhat lower than the low deposition ratios immediately north of Lake Ontario. The smaller spatial variability of the deposition ratios compared with the emission ratios near large point sources is probably related to a rapid rate of atmospheric mixing relative to the rates of wet-deposition processes. Alternatively, a preferential dry deposition of SO_2 relative to that of NO_2 may occur in the near-source region, or the differences may indeed represent a suppression of SO_2 oxidation within a short distance from the emission sources. However, the general similarity of wet-deposition and emission ratios on an annual average basis suggests that the incorporation of sulfuric acid in rain is not retarded with respect to the conversion and incorporation of nitric acid in rain.
>
> A small body of data suggests that the historical trends in the molar deposition ratios of sulfate to nitrate correspond to the trend in molar SO_2/NO_x emission ratios. The historical trends of the molar deposition ratio $SO_4^=/NO_3^-$ at Hubbard Brook and the molar ratios of SO_2 to NO_x in emissions in the eastern United States by EPA Region are plotted in Figure 4.19. Also shown in the figure is the result of a linear-regression analysis of the Hubbard Brook deposition ratio against time. Between 1965-1966 and 1979-1980, the linear regression gives a slope of -0.039 (standard error 0.006) and an intercept of 1.444 (standard error 0.054). The significance level of the slope is 0.0001.

Another perspective is provided by comparing the annually averaged regional wet-deposition rates with regional emissions. Integrating wet deposition over the northeast quadrant of the United States [using the isopleths of Pack (1980) based on MAP3S and SURE data] Golomb (1983) found sulfate deposition to be 4.3×10^{10} (±20 percent) moles/year and total nitrate deposition to be 3.9×10^{10} (±25 percent) moles/year. Golomb calculated regional emissions from U.S./Canada (1981) to be 2.2×10^{11} moles/year of SO_2 and 1.8×10^{11} moles/year of NO_x. The emissions were similarly distributed geographically. Golomb also indicates that 19 ± 6 percent of the sulfur and 20 ± 7 percent of the nitrogen emitted in this region are wet deposited there. It is interesting to note that Golomb found the molar ratio of sulfate to nitrate wet deposited annually in this region to be 1.1, compared with an annually averaged molar ratio of emissions for SO_2 and NO_x of 1.2. The similarity of the ratios suggests that, whatever specific processes are taking place, there is a tendency on an annual basis for sulfate and nitrate to be deposited in the northeast quadrant of the United States with no significant loss of one component relative to the other.

When Reviewer D realizes that atmospheric mixing may be important, he resorts to simplistic hypothetical considerations of transport which are relatively meaningless.

Reviewer D is concerned with the large discrepancy between emissions and deposition ratios in the northern Canadian provinces. However, these large emission ratios (e.g., 30.96 for northern Quebec) are associated with only 560×10^{-3} tons of SO_2 per year in a region of about 480,000 square miles. In terms of average annual emissions of SO_2 per unit area, the northern Quebec region resembles Nebraska. The report restricted its comments regarding ratios to the region bounded on the north by the acidity isopleth of pH = 4.6. At lower acidities, it is not possible to discriminate between the effects of regional anthropogenic sources, imported sources, and natural sources on the $SO_4^=/NO_3^-$ ratio. The report maintains only that within the region of precipi-

tation acidity higher than 4.6, i.e., clearly anthropogenic, there is no evidence of large scale nonlinearities.

Reviewer D maintains that an insignificant correlation exists between weekly $SO_4^=/NO_3^-$ ratios at Hubbard Brook and SO_2/NO_x emissions in Federal Regions I and II. It is not statistically feasible to compare weekly deposition ratios with annual emission ratios. If reviewer D adjusted the weekly values to obtain annual averages as he did in the case of sulfate concentrations, the results are of questionable relability.

Review E has written his comments with such an extraordinary degree of rancorous disdain that we are tempted to neglect his comments completely. The reviewer must have been driven by desperation to resort to such levels of distorted analysis. His approach is typified by the following salient argument copied from his review:

12. pp. 127-131. Here's what I want you to plot. Take the molar ratios for Europe (given in Table 4.7) and plot them against distance, say from your Region 1, and then add the data for N. America (your Fig. 4.17, p.130). Sure, it'll be crude:

Station	Distance (deg. lat.)	Ratio (normalized)
Europe:		
1	0	1
2	4	1.2 or .7
3	5	1.2 or 1.3
4	8	1.8 or 1.3
5	14	2.0 or 3.8
N. America:		
PM	0	1
Adirondacks	2	1.2
Quebec City (Your 1.6)	5	1.6

OKAY, NOW PLOT THEM. NOW, WHAT WERE YOU GUYS SAYING ABOUT CONSTANT MOLAR RATIOS??????????? This is so bad that even if I weren't out of time, I don't know how I could continue. Pure unsubstantiated bull! The opposite conclusion to theirs is staring them in the face!

Region I was selected by Rodhe et al. to represent the industrial source region, and in fact comprises the Ruhr Valley and the intensively industrialized area to the West of it. The other regions were chosen to represent downwind areas in which the average chemistry of the precipitation would reflect changes in the chemistry of the pollution plume as a function of distance from the source region. It is assumed that pollution sources enroute are not large enough to introduce large changes in the plume chemistry. The NRC acid deposition report examines the molar ratio of sulfate to nitrate in each of the regions to discern whether rapid consumption of oxidant by nitrogen oxides in the source region retards the oxidation of SO_2 until the plume travels to greater distances. The observed ratio increases with distance consistently with this hypothesis, but as the report states, there are other explanations. The report states however that the lack of a gradient in ratios is not consistent with the hypothesis, and in North America no gradient is apparent.

Reviewer E chooses to normalize the ratio of sulfate to nitrate to unity at the source of the region with the simple proportion: X/region ratio = 1.0/source ratio. However, it is clear from Figure 4.17 that the ratio is not increasing between Pennsylvania and the Adirondaks, and his normalized ratio should be 1.0 for the Adirondaks, not 1.2 as he indicates. The nearest station to Quebec City indicates an observed average of 1.3, or a normalized value of 1.2, not 1.6 as Reviewer E indicates.

A more serious distortion, however, arises from reviewer E's use of central Pennsylvania as the principal origin of a regional plume causing acid rain over North America. A more realistic comparison with the source region selected by Rodhe et al. would be the Ohio River Valley. In the attached

copy of the NRC acid deposition report Figure 4.17, arcs are drawn from Cincinnati (a central location in the Ohio River Valley) corresponding to the linear distances between the center of each downwind region and the source region in the European map (Figure 4.16). Contrary to the table of reviewer E, the figure indicates that even at 8 degrees distance from the source region there is no indication of an increasing gradient of the ratio.

The NRC acid deposition report is limited to a discussion of $SO_4^{=}/NO_3^{-}$ ratios to regions where the pH is 4.6 or lower, since at higher pH values the ratio is probably influenced at least as much by local SO_2 sources, natural sources, or globally distributed, anthropogenic sources as by long range transport from the Ohio River Valley. At pH 5.0 the acidity is within the range of global background values (NRC acid deposition report, p. 13). The northernmost region in the European map was characterized by a $SO_4^{=}/NO_3^{-}$ molar ratio of 5.7, and was also typified by a pH of 4.6 in 1974 (Likens et al., 1979). However the analogous outermost arc on the attached version of Figure 4.17 lies at 1,150 miles from the source region. It falls in the region of pH 5.0 or higher in North America, and the ratio can no longer be realistically considered as reflecting principally chemical transformations in the original plume.

Referee E has implied that the gas phase chemistry is unimportant in acid deposition in northeastern North America. Sulfuric acid generation occurs in clouds and in gas phase reactions. Indeed evidence suggests that oxidation of sulfur dioxide in clouds in northeastern North America may be more important than that occurring in the gas phase. However the good oxidants, ozone, hydrogen peroxide, and possibly other peroxy compounds, which are believed to be the necessary reactants for sulfur dioxide (bisulfite ion) in clouds, all are formed in gas phase chemistry. Thus in terms of current theory for clouds which have low concentrations of metal ion catalysts (Fe^{3+}, Mn^{2+}, etc.), if we don't

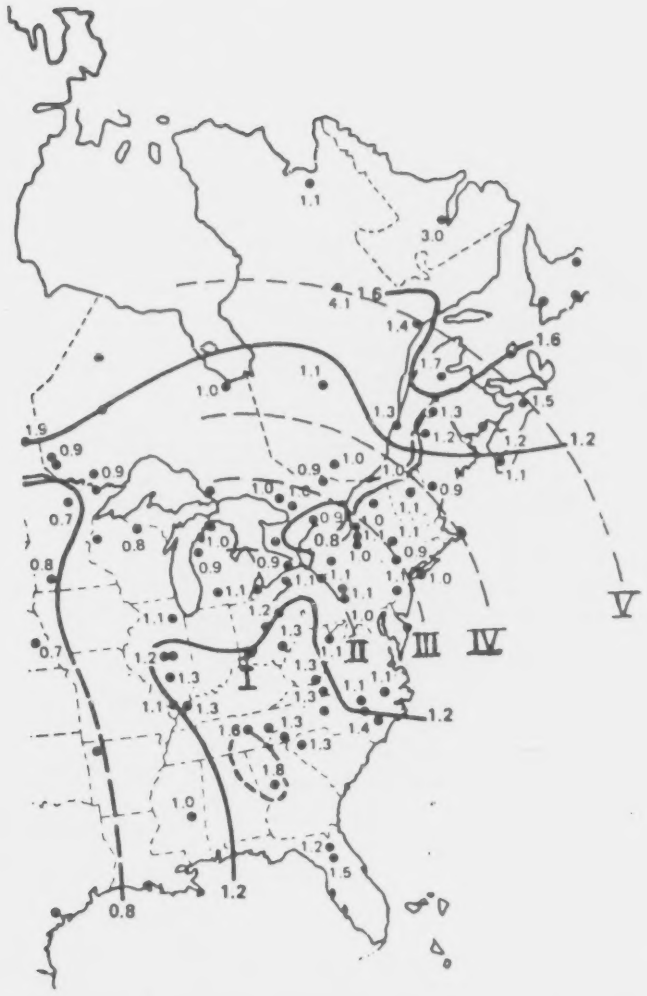

FIGURE 4.17 Average molar ratio of sulfate to nitrate in precipitation in eastern North America in 1980. SOURCE: B. Heikes, National Center for Atmospheric Research, personal communication (1983). Based on data from U.S./Canada Work Group #2 (1982). (Arcs drawn in the figure have been added to show the distance relationships which correspond to those used in the European map; Fig. 4-16.)

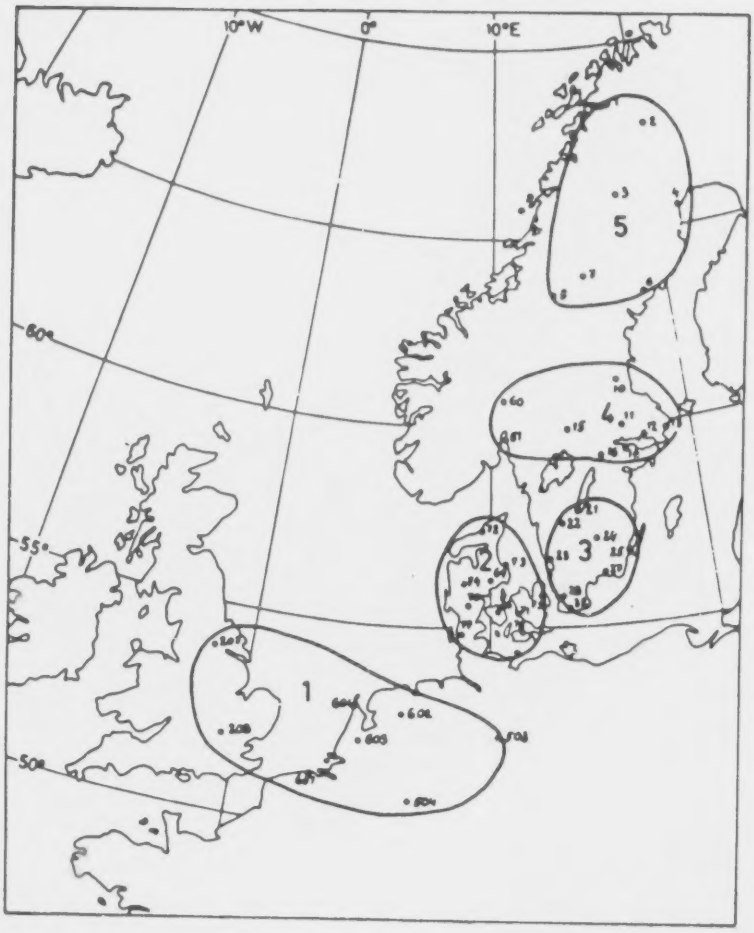

FIGURE 4.16 Locations of monitoring stations used in the data analysis of Table 4.7.
SOURCE: Rodhe et al. (1981).

include gas phase chemistry we cannot account for the rapid rates of acid generation which have been observed to occur in clouds. Gas phase and solution phase reactions are highly coupled, and no scientist today would attempt to rationalize the chemistry involved in acid deposition without including both gas phase and solution phase processes.

One other important conclusion of our NRC acid deposition report has been been questioned. We conclude that the reliability of long-range transport and transformation models is presently low in relating specific sources with receptor sites in sensitive areas. This conclusion does not seem to differ from that expressed in the report of the Interagency Task Force on Acid Rain (with which the Manowitz group has some undefined relationship), and the brief report of the Acid Rain Peer Review Panel. It also appears to be in accord with the comments of reviewer C (Bruce B. Hicks) of the newly added Oak Ridge "review" group. Work of Dr. Jack Shannon of Argonne National Laboratories has shown in very recent unpublished simulations using the ASTRAP model (private communication to Jack G. Calvert) that very good correlations are found for the calculated annual wet sulfate and nitrate deposition in eastern North America and the depositions observed in the 100 MAP3S, NADP, CANSAP, and APN sites for the year 1980. See the communication from Dr. Shannon in the attached letters in Appendix D. Such results look encouraging as related to long-time averages over large area. It is interesting that a proportional or linear chemical model (with diurnally and seasonally varied rates of SO_2 oxidation), rejected by the Manowitz group, was employed in these seemingly successful long-term average simulations. A more important question at hand is how well present models can do in establishing the percentage of the deposition in a given sensitive area which can be ascribed to a given emission source and

the extent of cutback in acid deposition in sensitive areas which we can expect from control of certain large sources which are well-removed from the site. It is the opinion of the NRC committee on acid deposition, the Interagency Task Force on Acid Rain, as well as the President's Panel on acid rain that this cannot be done now with confidence. Models are used by all of us in the sciences today, and certainly definitive uses have been and will continue to be made of them. Our caution relates to their inappropriate use. If such modelling is used in control policy formulation at the present time, and, of course it is being used now, then the modeller should provide the user with realistic estimates of the magnitude of the uncertainty associated with the results so that these can be incorporated into the control strategy planning. Such a practice is not suggested to frustrate the control plan, but it is necessary to provide the sense of realism that is demanded in view of the very high costs involved in the contol of sulfur oxide emissions.

One final point should be made. The NRC report states that the Rodhe, Crutzen, Vanderpol model (1981) which was used to explain the possible nonlinearity between $SO_4^=$ deposition and SO_2 emissions should be modified in the light of recent specific laboratory findings (Stockwell and Calvert, 1983). Reviewer A doubts that this modification would significantly influence the result. It should be noted that the suggestion of the NRC report, supported by simulations of Professor Perry Samson of the University of Michigan which are given in the report, was tested independently in a comprehensive model of the gas phase chemistry of SO_2, NO_x and hydrocarbons by Hov (1983). It was found that complete regeneration of radicals in the SO_2 oxidation reactions as suggested by the laboratory study leads to a nearly proportionate (linear) response of reduced sulfuric acid formation to reduction in SO_2 emissions in long-range transport; 50% reduction in SO_2 gave a 48% reduction in sulfate generated.

Certainly the conclusions of the NRC committee on acid deposition are necessarily very limited in nature, and indeed they must and will be checked by extensive further experimentation. Our conclusions represent the best judgment of a group of highly qualified and diversified scientists, and we certainly would not alter or compromise these conclusions on the basis of the unusual "review" of the Manowitz group.

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Attached in the next seven pages of this report is a discussion presented by Professor James Williams of the Department of Statistics, Colorado State University related to the statistical problems in treating the Hubbard Brook data.

References:

Hov, Ø. (1983), "Model Calculations of the Effect of SO_2, HO_x or HC Emissions Changes on Regional Scale Sulfate and Nitrate Production," p. 214-219 in Proceedings of the CEC Workshop Organized as Part of the Concerted Action "Physical Chemical Behavior of Atmospheric Pollutant", ed. S. Beilke and A. J. Elshout, D. Ridel Publishing Co.

Likens, G. E., P. F. Wright, J. N. Galloway, and T. J. Butler (1979), Acid Rain, Scientific American, 241, 43-51.

Munn, R. E., et al., (1982), Final Report to the U. S. Department of Energy, A Climatological Analysis of the Hubbard Brook (New Hampshire) Precipitation Chemistry Data, Grant DE-F G01-SOEV-10455.

Rodhe, H., P. Crutzen, and A. Vanderpol (1981), Formation of Sulfuric Acid and Nitric Acid in the Atmosphere During Long-Range Transport, Tellus, 33, 132-141.

Stockwell, W. R., and J. G. Calvert, (1983), The Mechanism of the HO-SO_2 Reaction, Atmospheric Environment, in press.

Tanner, R. L., and B. P. Leaderer (1982), Seasonal Variations in the Composition of Ambient sulfur-containing aerosols in the New York Area, Atmospheric Environment, 16, 569-580.

Comments on the "Review" of Manowitz reference D by Professor James S. Williams, Department of Statistics, Colorado State University, Fort Collins

A. Linear and Nonlinear Systems; Linear and Nonlinear Regression

Members of the National Academy of Sciences study panel have focused on the question of whether or not emission-deposition processes for SO_2-SO_4 and NO_x-NO_3 are linear or nonlinear. Reviewer D contests the published conclusion by panel members that linearity is a workable approximation for those systems in North America that are like the one sample in the Hubbard Brook Experimental Forest in New Hampshire. The definition of linearity set out in the panel report (p. 73) appears to be the same one used by the reviewer in much of his analysis, discussion, and conclusions. However the definition is too restrictive, as noted (at least by implication) in both the report and review.

A linear system in real variables is one in which input variable $\underline{x}$ is transformed into $t(\underline{x})$ where the function $t(\cdot)$ is characterized by:

$$t(\underline{x} + \underline{y}) = t(\underline{x}) + t(\underline{y}) \text{ for all } \underline{x}, \underline{y} \in R^p$$

In such a system if input is changed from $\underline{x}$ to $m\underline{x}$, then output is changed from $t(\underline{x})$ to $mt(\underline{x})$. In particular if $\underline{x}$ is a numerical measure of man-made emissions and $t(\underline{x})$ is a measure of depositions, then reducing emissions to a fraction m of $\underline{x}$ would reduce depositions to the same fraction m of $t(\underline{x})$. If man made emissions were eliminated ($m = 0$), then depositions would be eliminated. This last result is unrealistic because undoubtedly there are depositions which are not the result of man-made emissions. This objection can be removed by considering other types of systems. Systems of the type just described are called homogeneous linear; a more appropriate type for consideration is nonhomogeneous linear systems for which:

$$t(\underline{x}) = t(0) + u(\underline{x})$$

$$u(\underline{x} + \underline{y}) = u(\underline{x}) + u(\underline{y}) \text{ for all } \underline{x}, \underline{y} \in R^p$$

In such a system input results in a homogeneous linear change added to output associated with zero input. If input is changed from $\underline{x}$ to $m\underline{x}$, then the output is changed from $t(\underline{x})$ to $t(x) + mu(\underline{x})$. In such a model for an emission-deposition process, the elimination of man-made emissions ($m = 0$) would not eliminate depositions, but would reduce them to a natural level ($t(\underline{0})$). Also the rate of change in output with respect to any particular input variable can be a constant other than 1, the only value it can be if the system is homogeneous linear.

A system which is not linear is nonlinear. It is characterized by rates of change which are not constants but instead depend on $\underline{x}$. For such a system, the change in output in response to a change in input can be calculated but it is more complicated in form than the change in output for a linear system.

The terms linear and nonlinear are also used to qualify different types of regression analyses. However, they are not used in this context in the same way that they are used to characterize a transformation of input into output. For example, the nonlinear transformation of a single input variable $\underline{x}$ into output $t(\underline{x})$ which is given by:

$$t(\underline{x}) = \beta_0 + \beta_1 x + \beta_2(1/\underline{x})$$

might be used as the basis of the model of linear multiple regression analysis. With respect to the regression analysis, linear applies to the manner in which the coefficients β_0, β_1, and β_2 enter the equation for $t(\underline{x})$ rather than to the manner in which $\underline{x}$ enters the equation. In general, the methods of linear regression apply to all linear transformations of input x into output $t(\underline{x})$ and either exactly or approximately to a very large class of nonlinear transformations. There are also methods of nonlinear regression analysis which can be applied to nonlinear transformations to replace approximate

procedures by exact ones and to enlarge the class of transformations of input to output that can be studied with a regression analysis.

The conclusion to be drawn from this discussion is that the conclusion presented by the panel members and contested by the reviewer is not as central as the former indicate and is not as restrictive to consideration of emissions control policy as claimed by the latter.

B. Statistical Analysis of Relationship of Deposition Data from Hubbard Brook Experimental Forest to Regional Emissions Data

Efforts to derive equations for the transformation of emission variables into deposition variables have not been successful enough to predict the latter in any given region from measurements of the former in nearby and more distant regions. One can predict with confidence that such equations will never be found because the system includes a number of random components which cannot be included in such equations. The successes of studies of this type will increase our knowledge of possible workings of tranport, chemical conversion, and deposition processes in the system.

Satisfactory predictions can be derived from the results of regression analysis if the more important variables affecting deposition can be identified and measured and if sufficient data can be collected to insure accurate results of a regression analysis. At the present time the complete list of important variables for even one deposition site, such as the Hubbard Brook Forest, is unknown, and only a modest amount of data are available for analysis. These two reservations are clearly stated in the panel report and are given as reasons for their basing conclusions on an informal analysis for qualitative descriptions of relationships. No attempts were made at formal analyses for quantitative descriptions and predictions.

The main difficulty one would encounter in a regression analysis of the relationship of the Hubbard Brook Forest data to the emissions data for the Mississippi valley and regions to the east can be seen in Figures 4.14 and 4.15 of the panel report. The modest trends present in the series of yearly deposition figures for $SO_4^=$, NO_3^-, H^+, and pH (Figure 4.14) are hypothesized to be responses to trends in the series of yearly emissions for five regions either nearby the Hubbard Brook Forest (Regions I and II) or more distant (Regions III, IV, and V). These emission trends contain strikingly different changes in direction for SO_2 and leveling out for NO_x. In each region there is a year of maximum SO_2 followed by a decline in levels. This occurs in 1965 for Regions II and V, in 1970 for Regions I and III, and in 1975 for Region V (Figure 4.15). The apparent decline in $SO_4^=$ deposit begins in 1968 (Figure 4.14). If it is a positive response to changes in emissions, then it must be a delayed effect of the turn downward in SO_2 emissions in Region II and/or Region V. If it is a negative response to changes in emissions, then it must be an indication of increasing dominance of Region IV on the level of SO_2 contamination reaching the Hubbard Brook Forest. In each region, the level of NO_x emissions either levels off or even declines slightly after a period of increase. This occurs in 1965 for Region III, in 1970 for Regions I, II, and V, and in 1975 for Region IV (Figure 4.15). The increase in NO_3^- deposition lasts until 1970 (Figure (4.14) and, therefore, is apparently a positive response to emission changes in Regions I, II, and V or a negative response to increasing dominance of Region IV on levels of NO_x contamination reaching the Hubbard Brook Forest. Also the possibility that effects on one source of acid deposition might be indirect, that is the consequence of changes in the other source, cannot be overlooked.

Summary indices of emissions from all sources, such as the totals for SO_2 and NO_x graphed in Figure 4.15 or the weighted totals for SO_2 and NO_x presented by Reviewer D, are inappropiate input variables unless they are constructed to emphasize by correct weighting, the real effects of the regions on the levels of contaminants in the air mass of the region where deposition is measured. Reviewer D has implied that weighting by either inverse distance or inverse squared distance is correct. However he presents no analysis to support this conclusion and his citation of the study published by Munn et al. is inadequate because the analyses prepared by that group are so few in number and unfocused.

An important point to realize is that the regression analysis cannot be used to identify sources of emissions which truly affect deposition variables in a region. However the analysis can be used to eliminate some of the regions which have negligible effects if the data record includes patterns in changes in emission trends which can be used to distinguish each region from all others. It is fairly certain that a long record would be needed to achieve this. It cannot be done with the available data because trends in Regions I, II, and III are similar for SO_2 and trends in Regions I, II, III, and V are similar for NO_x emissions (Figure 4.14).

Reviewer D has based his conclusion that yearly SO_4 depositions are not linearly related to SO_2 emissions on regression analyses in which an input index is used. An example of one of these indices is graphed in his Figure 2 along with a graph of adjusted SO_4 depositions in the Hubbard Brook Forest. The reason that there is no significant linear relationship is obviously due to the marked counter trends in the two series up to 1970. Were Region II to receive greater weighting in the index, much of the upward trend in the SO_2 trace would be eliminated, and a stronger positive linear relationship would

be obtained. Were Region IV to recieve greater weighting in the index, much of the downward trend in the SO_2 trace would be eliminated, and a strong negative linear relationship would be obtained. It is clear that the decision of how to weight regional emissions in the index determines the sign and the statistical significance of linear dependence of SO_4 depositions on SO_2 emissions. Were the correct weightings known, the regression analysis could be properly applied to determine if there is evidence in the data for linear dependence. Panel members in their report have argued that the correct weightings are not known and that as a consequence a formal analysis is not justified (p. 140-141).

C. Statistical Adjustment of Deposition Data to Eliminate Effects of Meteorological Variables

Reviewer D has used linear regression procedures to adjust deposition measurements taken at Hubbard Brook Forest for precipitation volume, surface temperature (at Concord, New Hampshire) and all other factors associated with cosine variation in seasonal trends. It is not clear whether the analysis for coefficients used in the adjustment was calculated from the data broken into yearly subsets (a within-year analysis) or from the unpartitioned records because the reviewer notes that coefficients are the averages of several coefficients (page 8). Presumably this comment refers to something else because only the latter method of calculation is appropriate.

The choice of cosine variable to represent cyclic trends is ineffective because year-to-year variations in seasonal effects which are necessary for a proper adjustment are not included in the cosine variable. The only consequence of inclusion of the cosine term could have would be to increase the residual variance of the adjusted records and thereby reduce the statistical significance of tests based on the adjusted data.

Both panel members and the reviewer note that summer deposition of $SO_4^=$ dominates the yearly record. Because of this, further analyses of $SO_4^=$ data should be done separately for the yearly records of depositions during warm and during cool seasons. Changes in the yearly deposition trend should show up more clearly in the warm season record than in the yearly record.

APPPENDIX D: THE ORIGINAL "MANOWITZ REVIEWS" OF THE NRC ACID DEPOSITION DOCUMENT*

*These "reviews were subsequently rewritten in part and submitted to the Honorable Don Fuqua, James H. Scheuer, and Marilyn Lloyd for their consideration. Since the attached "reviews" were circulated widely within the Power Industry and Government circles and Jack G. Calvert did not have access to the revised document until a few days before these comments were due in Washington, Calvert's response to the "reviews" is based upon the originals which were sent to him. The various anonymous reviews which appear here can in large part be identified through the new document. These are as follows:

Reviewer A: Stephen E. Schwartz (Brookhaven)

Reviewer B: Lawrence I. Kleinman (Brookhaven)

Reviewer C: Paul Michael (Brookhaven)

Reviewer D: Fred Lipfert (Brookhaven)

Reviewer E: Not identified, but the highly edited version of this is attributed to W. G. N. Slinn, M. T. Dana, and R. C. Easter (Battelle Pacific Northwest)

Reviewer F: Largely due to Jack Shannon (Argonne)

The scientific questions raised in the reviews attached are essentially those in the revised "reviews" which were submitted to the House Committee on Science and Technology by Bernard Manowitz of Brookhaven National Laboratories recently.

BROOKHAVEN NATIONAL LABORATORY
ASSOCIATED UNIVERSITIES, INC.
Upton, Long Island, New York 11973

Department of Applied Science (516) 282- 3037
FTS 66-

July 26, 1983

Dr. Herbert Friedman, Chairman
Commission on Physical Sciences,
Mathematics and Resources
National Research Council
2101 Constitution Avenue, N.W.
Washington, DC 20418

Dear Dr. Friedman:

As Chairman of the National Laboratory Consortium of the Interagency Task Force on Acid Precipitation, I have been asked by two federal agencies and by a Congressional committee to solicit comments from the National Laboratories on the document recently issued by the National Academy Press entitled "Acid Deposition Atmospheric Processes in Eastern North America" under your aegis.

Since I understand that your Commission is meeting on August 1, 1983 and since the reviews I have received thus far disagree with the major findings of the Academy report, I am bringing a series of these reviews to your attention.

The major points made by the reviewers are these:

- The historical record of wet deposition at Hubbard Brook, NH, in relation to emissions trends in the Northeast, plays a major role in the arguments for "linearity" (and implied proportionality). Since Hubbard Brook is well outside the region of maximum emission and deposition, it is unlikely that experimental data could reveal whether processes are linear or not. (Reviewers C, D).

- There are strong arguments on theoretical grounds that non-linear atmospheric chemistry processes can and do occur. Such non-linearities would result in a less than 1.1 proportionality between emission and deposition. (Reviewers A, B, E).

- The Academy report attempts to relate the deposition trends at Hubbard Brook to Northeastern emission trends in a very superficial way, without confidence limits on the findings. Detailed statistical reanalysis of Hubbard Brook data, using multiple regression analysis to account for meteorological variability over the period of record as well as emission estimates for every year, finds a substantially less than 1:1 proportionality with a high degree of confidence. (Reviewers D, E, F).

In view of the negative and substantive comments of a significant number of reviewers, all of whom are active in the field, I question the quality of the review given to the Academy report.

Sincerely,

Bernard Manowitz
Chairman - National Laboratory
Consortium of the Interagency Task
Force on Acid Precipitation

BM:mh
att. 6
cc: D. Beecy, Chairman, Task Group B
O. Bentley, Co-Chair, ITF
J. Calvert, Chairman, NRC Comm.
K. Demerjian, Chairman, Task Group C
R. Kasper, Exec. Dir., Comm. Phys. Sci., Math. & Resources
L. Machta, Co-Chair, ITF
C. Riordan, Co-Chair, ITF

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01 AUG 1983

July 26, 1983

Dr. Raphael G. Kasper
Executive Director
Commission on Physical Sciences,
Mathematics and Resources
National Research Council
2101 Constitution Ave., N.W.
Washington, DC 20418

Dear Dr. Kasper:

I recently had a conversation with Dr. Herbert Friedman, Chairman of the Commission, regarding a number of negative comments I have received on the Academy's Acid Deposition Report. He asked that you make the attached letter and comments available for the Commission's August 1, 1983 meeting.

Sincerely,

Bernard Manowitz

Bernard Manowitz
Chairman, National Laboratory
Consortium of the Interagency
Task Force on Acid Precipitation

pt
att. 7

28-916 0 - 84 - 48

Reviewer A

Review of National Academy Report on Acid Deposition

This report focuses upon the relationships between emissions of acid precursors and the deposition of acidic substances. The stated purpose is "to assess the current state of scientific information that can be marshaled to describe those relationships in the hope that our assessment will be valuable to decision makers in the government and in the private sector" (pp. 1-2).

Overall, the report exhibits a number of positive qualities, but in my judgment it is seriously flawed. On the positive side, it delineates well the scientific issues that confront the regulatory sector. The red herring of natural sources is readily dispensed with (p. 3). The scientific issue is thus drawn as establishing the relationships between anthropogenic emissions and deposition. The report also defines the sub-issues of nonlinearity and distant vs. local sources. The report draws the conclusion that "evidence exists for long-range transport of pollutants leading to acid deposition", but cautions that "the relative contributions of specific source regions to specific receptor sites currently remain unknown" (p. 5). The report points out gaps in knowledge requisite to establishing such contributions and suggests pertinent laboratory, field, and modeling studies to better define such relationships. The report concludes that at the present level of understanding "we cannot objectively predict the consequences for deposition in ecologically sensitive areas of changing the spatial pattern of emissions in eastern North America, such as by reducing emissions in one area by a larger percentage than in other areas" (p. 11). In my judgment, the report makes an adequate case for the above conclusions. However, as I argue below, the report might well have offered some guidance, particularly with respect to the latter question.

On the negative side, I submit that the report is deficient insofar as it focuses, almost entirely, on long-term average (e.g., annual) deposition, to the exclusion of consideration of information that has been gained from shorter-term measurements. While such a focus may be appropriate from the perspective of ecological concerns and/or from a regulatory perspective, a wealth of detail is lost that is pertinent to the mechanism of acid deposition, and pertinent, therefore, to the stated subject of the report. The focus on long-term relationships between emissions and deposition leads to a number of conclusions that either are erroneous or subject to misinterpretation or, alternatively, are so narrowly drawn as to border upon the self-evident. I call particular attention to the following:

(a) "Pollutants are being thoroughly mixed over a region with linear dimensions up to 1000 km" (p. 9). This conclusion derives from the claimed near-constancy (in space and time) of the ratio of long-term average sulfate and nitrate concentrations in precipitation and the near equality of this ratio to the ratio of the oxides in emissions. The statement is patently incorrect if we take "thoroughly mixed" in the usual sense of "exhibiting uniform concentration over the volume in question, at a given instant or

over a given period of time." Evidently, the authors mean simply to say that the ratio of long-term average atmospheric concentrations is uniform spatially. In any event, the statement as it stands is erroneous and certainly detrimental to conveying the authors' point.

(b) "There is no evidence for strong nonlinearity in the relationships between long-term average emissions and deposition" (p. 7). Presumably the authors mean to say here, "There is no evidence for strong nonlinearity in the relationships between long-term average emissions and long-term average deposition." Accepting that, the statement summarizes the search for such a relationship in available, long-term records of precipitation concentration. As it turns out, there is essentially only one station that meets the authors' criteria for such records of acceptable quality, and the authors claim to discern no such relationship for that station. While that may be argued (and I am sure it will be), restricting the discussion to long-term averages excludes a wealth of evidence from short-term measurements (event or sub-event) that is demonstrative of mechanistic nonlinearities, the understanding of which is crucial to the description of the source-receptor relationship in acid deposition.

(c) "If we assume that all other factors, including meteorology, remain unchanged, the annual average concentration of sulfate in precipitation at a given site should be reduced in proportion to a reduction in SO_2 and sulfate transported to that site from a source or region of sources" (p. 10). This statement is so highly restrictive as to be essentially useless. Strictly, it deals only with the relationship between annual average concentration of sulfate in precipitation at a given "site" and, presumably, annual average atmospheric SO_2 and sulfate at that "site," under the assumption that "all other factors, including meteorology, remain unchanged." Even granting the validity of the statement as it stands, the statement is all the more important for what it does not treat, i.e., (1) the relationship between emissions and annual average atmospheric concentrations at sites remote from emissions, and (2) the factors which are assumed to remain unchanged. While it might be hard to quarrel, on the basis of annual-average measurements, with the statement as it stands (although I can think of at least three mechanistic reasons why it might not be expected to hold in the short term and can cite, as well, any number of studies to that effect), I am concerned that the statement may be misconstrued as relating deposition to emissions, particularly insofar as it appears under the heading "Implications for Emission-Control Strategies," and immediately follows the statement that "analysis of observational data provides guidance for assessing the consequences of changing SO_2 emissions for wet deposition of sulfate."

(d) "Assuming that all other emissions and conditions remain unchanged, we would expect that if the molar ratio [of SO_2 to NO_x] in emissions in eastern North America were changed by changing SO_2 emissions, a similar change would occur in the ratio of sulfate to nitrate in wet deposition" (p. 10). The authors then go on to qualify the statement, indicating that "because the analysis is based on spatial distributions, its applicability is limited to circumstances in which the spatial distribution of emissions is not changed." Here, as above, I am concerned about possible

miscontruction of the intent of the authors' statement. I am sure that some will see in this statement a policy implication that if sulfur oxide emissions are to be reduced, they should be reduced in such a way as to preserve the existing spatial distribution. On the other hand, I would foresee others who would take statement (a) above, that "pollutants are being thoroughly mixed over a region with linear dimension up to 1000 km," as suggesting that reduction in SO_2 emissions anywhere within the region will be equally effective in reducing annual average deposition of sulfate at all other points in the region. I submit that the proper strategy for emissions reduction to protect ecologically sensitive regions lies somewhere between these two extremes. Unfortunately, the report provides very little guidance to the policy makers who must formulate such a strategy.

In addition to the foregoing general comments, I have a number of comments relative to the linearity-nonlinearity issue. But first we must examine why linearity-nonlinearity is of interest at all. Clearly, if the geographical region is taken sufficiently large, then the long-term total deposition of SO_2 and $SO_4^=$ integrated over this area is linear in emission of SO_2 in this area, and for that matter equal. The question of nonlinearity in long-term deposition is therefore inherently a question of whether the scale of transport from source to receptor is influenced by the emission source strength. Clearly also, any negative departure from linearity in a given receptor region must be compensated by a positive departure from linearity in some other receptor region or regions. Thus, it is evident that the entire question of linearity-nonlinearity is deeply imbedded in the larger question of the scale of transport of the pollutants of concern. This point is not well brought out in the report. The failure to appreciate this point may be responsible for some of the confusion in the report over the question of nonlinearity.

Some specific comments on the treatment on nonlinearity follow.

(1) For all the care given to the definition of linearity (pp. 72-75), the report still seems to confuse linearity with proportionality. Thus, (p. 116): "The central question is whether the data [obtained from monitoring networks] can be used to judge whether deposition is linearly or nonlinearly related to emissions.... If the relationship is linear, a change in emissions would be reflected in a proportionate change in deposition; if it is nonlinear, the change in deposition, if any, would be disproportionate." The second statement above is at best loosely phrased and at worst flat out wrong. It could be corrected as follows: "If the relationship is linear, a change in emissions from a given source (or set of sources) would be reflected in a proportionate change in deposition due to that source (or set of sources)," and similarly for the second clause. The addition of the logical connection between the source and the receptor is, in my judgment, central to the entire question of acid deposition, and ultimately to the question of control strategy. However, setting that aside for the moment, it is disappointing to say the least to see propagation of such confusion in this forum.

Going beyond definitions, we see that the foregoing misconception is carried over into the discussion of the model runs by Samson (p. 76). Here, calculations are compared with and without the contribution of a "background" source. The report states that the shift from the solid to the dashed sets of curves depicted in Figures 3.1 and 3.2 is "the result of eliminating the trivial source of nonlinearity arising from the background source, term B in the equation, c = kS + B." This result is indeed trivial but it is not a nonlinearity and in any event is an observation that should hardly warrant discussion. The only reason that such a discussion can possibly have persisted into the final report is an inherent lack of understanding of the linearity-nonlinearity concept.

One further amusing (or saddening) point is the passing reference in the same paragraph to Figure 3.2 depicting "a result that is essentially linear (with proportionality constants less than unity)." The point of figures such as 3.1 and 3.2 is that any departure from the one-to-one line of percent $\Delta SO_4^=$ concentration vs. percent ΔSO_2 (concentration or emission), sulfate concentrations from other, unchanged sources having first been subtracted out, is an inherent nonlinearity. In fact, the 24- and 48-hour graphs represent a departure from linearity of ca. 30%, potentially very important in the context of control strategies. This is not to argue that the model calculations are correct, but only that they are incorrectly interpreted.

(2) The report argues at some length (pp. 75-81) that model calculations of Rodhe et al. suggesting nonlinearity may be an artifact as a consequence of the incorporation in the model of a radical termination step involving SO_2, whereas the actual reaction may well be a chain propagation step: "The participation of reaction (3.1) results in a direct nonlinear feedback into the SO_2 oxidation mechanism, while reaction (3.2) does not seriously perturb the concentration of the hydroxy radical" (p. 78). This argument seems flawed by the following reasoning. We note (p. 80) that in the simulation studies of Calvert and Stockwell, only about 10% of $HO-HO_2$ radical termination occurs through the $HO-NO_2$ reaction, for the quite high NO_x concentration of 100 ppb (and less for lower NO_x; indeed, the report suggests a curious quadratic dependence). In any event, we note that the rate constant of the $OH-NO_2$ reaction is about tenfold that of the $OH-SO_2$ reaction, so that for 100 ppb SO_2, only 1% of radical termination would be due to SO_2 (again less for lower SO_2). Thus, the possiblity of any significant influence on radical concentration due to SO_2 termination reactions would appear to be ruled out.

(3) The discussion of nonlinearity in SO_2 deposition due to aqueous-phase processes (cloud, rain) is short (1.5 pages; pp. 81-82) and superficial. Reference is made essentially only to the H_2O_2 oxidation mechanism of SO_2 oxidation, focusing principally on the limiting reagent process. No discussion is given at all of the pH dependence of SO_2 solubility and aqueous reaction kinetics, which would be inherently nonlinear insofar as H_2SO_4 (and to a lesser extent SO_2 itself) would influence cloudwater pH. This process has been the subject of much research over the past decade (as summarized in Appendix A, pp. 178-193), but little of this research finds it way into the discussion in the text. Other types of processes leading to nonlinearity (e.g., non-constant nucleation efficiency as a function of aerosol loading) are also not treated.

In summary, the relationship of the linearity-nonlinearity issue to the larger problem of scale of transport and control strategies does not appear to be adequately recognized. Moreover, the details of the discussion of nonlinearity are superficial and flawed, and little recognition is given of the importance of the fact that nonlinearities, as exhibited in short-term phenomena, can influence the source receptor relationship. For these reasons, little import should be given to the conclusion of a lack of evidence for nonlinearity in the relationship between long-term average emissions and deposition.

Reviewer H

Comments on
"Acid Deposition - Atmospheric Processes in Eastern North America -
A Review of Current Scientific Understanding,"
National Research Council.

Three major issues are considered and appear to serve as foci for the material reviewed in the report. As stated on pages 6-9, these issues are (1) "applicability of models to decisions on control strategies," (2) "non-linearity" and, (3) "influences of local and distant sources."

I. Models:

The authors have applied a healthy degree of skepticism and insight in arriving at their conclusions regarding the usefulness of models. To paraphrase the report (pp 148-149) ..... a great deal of research is required before the acid deposition problem can be described with precision by models. It is fairly clear that in this statement the usefulness of models is judged relative to the needs of policy makers, not to the needs of research scientists. The use of models, even complicated ones, by the latter group, should be judged by different standards. Although models cannot be used to make a priori predictions of many of the important features of the acid deposition problem, they can be used to good advantage by the research community in designing experiments, interpreting data, performing controlled computer experiments on the environment and, in general, increasing our understanding of how the various pieces of a complex problem interact with each other.

II. Nonlinearity:

An unsupported conclusion is reached regarding the relation between emissions and deposition. "It is ..... improbable that the oxidation of SO_2 is sufficiently hindered by a lack of oxidant to cause a disproportionately small

reduction in sulfate concentrations as a result of a given reduction in SO_2 emissions." (p 9). The "evidence" in the text for this statement derives from a comparison of deposition at Hubbard Brook with emission, from consideration of the gas phase oxidation chemistry of SO_2 and from the sulfate to nitrate molar ratio in precipitation as compared with emissions. This evidence, some of which can be interpreted in different ways, as described below, does not support the stated conclusion that deposition is linearly related to emissions. At best, one could say, "Direct evidence of a strongly nonlinear relationship between wet deposition of sulfate and SO_2 emissions is limited to extensive historical data in Europe." (p 139). Specific comments on the "evidence" against nonlinearity follows:

- p 42, also p 84. "In theory, the amount of H_2O_2 formed in gas-phase reactions and taken up in cloud water and precipitation, is sufficient to oxidize a large fraction of S(IV)." Many photochemical calculations of the PBL predict a H_2O_2 concentration of a few tenths to approximately one ppb (Schiavone and Graedel, 1981; Rodhe, et al, 1981; Hov, 1983). In order to account for the composition of summertime precipitation in the northeastern U.S., several times this amount of oxidant and SO_2 is required. It is certainly possible that future calculations or experiments will show that $[H_2O_2] > [SO_2]$, perhaps as a result of an in-cloud or free tropospheric source of H_2O_2. It is misleading, however, to imply that a large enough oxidant supply has been identified.

- p 50. "The most important attachment process under most in-cloud conditions is undoubtedly nucleation." Judging from a following sentence, diffusional attachment (of gases and particles) appears to be a subset of "attachment."

Either there is a semantic difficulty or the role of gas phase SO_2 and HNO_3 in acidifying precipitation is seriously understated.

• p 80. The criticism of the nonlinearity produced in Rodhe's model is supported by many other experimental and theoretical studies (see, e.g., Miller, 1978; Hov and Isaksen, 1981). Although it appears likely that the gas phase oxidation of SO_2 is linear, there is no assurance that the aqueous phase oxidation is likewise linear. It is a deficiency of the main text (Chapters 1-5) that the relative amounts of gas and aqueous phase oxidation are not treated. Estimates of the relative importance of these processes have been made based upon the isotopic composition of precipitation (Holt, et al, 1981), the abundance of aerosol SO_4^{2-} (Newman, 1979; McNaughton and Scott, 1980), and a comparison of the acidity of precipitation and aerosol (Newman, 1979; Scott, 1982). At this point, the evidence, though largely inferential, points strongly to the in-cloud oxidation process as being the largest contributor of SO_4^{2-} to precipitation (Oppenheimer, 1983). The concern is that the aqueous pathway is the most significant and that it is nonlinear due to either pH inhibition or oxidant limitation.

• p 120-123. Comparison of Hubbard Brook data with emissions. It is misleading to base this comparison upon two years of monitoring and emissions data, namely 1965 and 1978. Emissions have a broad maximum around 1970 which is not reflected in precipitation composition. The two sets of curves, in fact, look sufficiently dissimilar such that one could argue that there is a complicated nonlinear relation between emissions and deposition.

• p 135. The NO_3^- to SO_4^{2-} ratios do not justify the conclusion that there is sufficient oxidant for all or most of the SO_2 within eastern North America. It admittedly is more straightforward to assume that equal deposition and emission ratios imply a high removal fraction for both substances than to arrive at an

explanation for why the average removal fractions are both equal but substantially less than one. The proposed high removal efficiency of NO_x is based upon the rapid formation of HNO_3 and the high solubility of HNO_3. A high NO_x wet removal efficiency is, however, not in accord with summertime, air quality observations in moderately polluted areas which indicate

$$\frac{SO_4^{2-}}{SO_2} > \frac{HNO_3 + NO_3^-}{NO_x}.$$

Perhaps HNO_3 is rapidly removed by dry deposition thereby making it unavailable for wet deposition. A NO_3^- to SO_4^{2-} molar ratio in precipitation equal to emissions could then occur without requiring efficient scavenging of S compounds.

• p 137. The data in Table 4.9 reflects the different average heights of NO_x and SO_2 emission sources. In general, during the nighttime $NO_x > SO_2$. This occurs, in part, because low level NO_x emissions are trapped above the boundary layer.

III. Source Attribution:

The text contains a good description of the monitoring evidence to the effect that receptor areas are impacted from many source regions, sometimes simultaneously. The complexity of the source attribution problem is conveyed as are the hopes and shortcomings of several diagnostic methods. The notion of strong mixing is, however, considerably overstated due to the authors' reliance on long term (monthly) average monitoring data.

Holt, B.D., Cunningham, D.T., and Kumar, R. (1981). Oxygen isotopy of atmospheric sulfates. Environ. Sci. Tech. 15, 804-808.

Hov, O. (1983). One-dimensional vertical model for ozone and other gases in the atmospheric boundary layer. Atmos. Environ. 17, 535-549.

Hov, O. and Isaksen, I.S.A. (1981). Generation of secondary pollutants in a power plant plume: A model study. Atmos. Environ. 15, 2367-2376.

McNaughton, D.J. and Scott, B.C. (1980). Modeling evidence of in-cloud transformation of sulfur dioxide to sulfate. JAPCA 30, 272-273.

Miller, D.F. (1978). Precursor effects on SO_2 oxidation. Atmos. Environ. 12, 273-280.

Newman, L. (1979). General considerations on how rainwater must obtain sulfate, nitrate, and acid. Presented at the Division of Environmental Chemistry, The Am. Chem. Soc., Honolulu, Hawaii, April 1979.

Oppenheimer, M. (1983). The relationship of sulfur emissions to sulfate in precipitation. Atmos. Environ. 17, 451-460.

Rodhe, H.P., Crutzen, P. and Vanderpol, A. (1981). Formation of sulfuric and nitric acid in the atmosphere during long range transport. Tellus 33, 132-141.

Schiavone, J.A. and Graedel, T.E. (1981). 2-D studies of the kinetic photochemistry of the urban troposphere. I. Air stagnation conditions. Atmos. Environ. 15, 163-176.

Scott, B.C. (1982). Predictions of in-cloud conversion rates of SO_2 and SO_4 based on a simple chemical and dynamic model. Atmos. Environ. 16, 1735-1752.

7/25/83
Review C

The Committee on Atmospheric Transport and Chemical Transformation in Acid Precipitation of the National Academy of Sciences in their recent report state that "there is no evidence for a strong nonlinearity between long-term average emissions and deposition," (Summary page 7). They admit that the conclusion is clouded by uncertainty (p. 9) due to "the limited amount and uncertain quality of the data, the natural variability of atmospheric processes, and the lack of firm understanding of the physical and chemical processes involved." Beyond these disclaimers, there are a number of questions that can be raised concerning the basis for the NAS conclusions. These points need resolution before the conclusions are used for the setting of emission control policies.

1. ONLY WIDE-AREA, LONG-TERM CONCLUSIONS ARE DRAWN

The NAS committee looked at long-term data only; they did not address the short-term (event) or the spatial scale of less than a few hundred kilometers. The million-square kilometer so-called mixing that they discuss occurs only as an artifact of averaging over long time periods.

On the other hand, it is, no doubt, true that if a large enough area is considered, the acid deposition response to emissions will be linear. The real question is "On what scale is the system linear?"

2. THE USE OF HUBBARD BROOK DATA IS QUESTIONABLE

The deposition concentrations at Hubbard Brook are somewhat lower than those in the region of highest concentrations (by almost a factor of two).

The very nature of mechanisms that can cause nonlinearities (i.e., limited amounts of oxidant, competition among species, pH-induced limitations on mass transfer, etc.) means that, at first approximation, the effect will be most evident where the concentrations are the highest. However, because of the fact that the system will appear linear if averaged over a large enough region, an opposite effect

is liable to be seen where lower concentrations occur far downwind of the high source and concentration regions. For example, a lack of sufficient oxidant in a region would allow unoxidized SO_2 to be vented from a storm and made available farther downwind. Hence, an increase (or decrease) in emissions might result in a smaller than proportional increase (or decrease) in concentrations near the region of high emissions; thus, a proportionally greater (or lesser) amount of unreacted primary pollutant will be available to be deposited downstream. Following this line of reasoning, it is quite possible that Hubbard Brook could be in a location where ambiquous results on linearity would be obtained. The attached figure illustrates the point.

3. INSUFFICIENT ATTENTION IS GIVEN TO IN-CLOUD CHEMICAL REACTIONS AS A POSSIBLE SOURCE OF NONLINEARITY.

There is a large amount of admittedly inferential evidence that a major portion of precipitation acidity derives from in-cloud oxidation. Quite a number of authors have put forth this point of view (Newman, 1979; Scott, 1982; Lazrus, et al, 1983; Oppenheimer, 1983; etc.). Hence, the effort to discredit the model of Rodhe, et al,(1981) might well be moot; the Rodhe treatment of in-cloud oxidation is quite simplified and does not seem to have been improved in the calculations reported in the NAS report. The report discusses how H_2O_2 production might be suppressed in a NO-rich polluted atmosphere and how it is unreacted SO_2. They recognize that this could be the source of nonlinearities, but note the lack of an adequate data base to test the possibility. The report states that the data "suggest that concentrations of (oxidant in cloud water) may be sufficient in eastern North America for complete oxidation of SO_2 except, perhaps, in winter." There are a number of photochemical calculations that would indicate that the H_2O_2 concentrations are several times too low. At best, the point is arguable; it might just be that the NAS report is incorrect.

4. "LACK OF EVIDENCE FOR STRONG NONLINEARITIES" IS NOT A PROOF OF LINEARITY.

Stating that "no evidence of strong linearity" could be found is far from being a logically sound proof of linearity.

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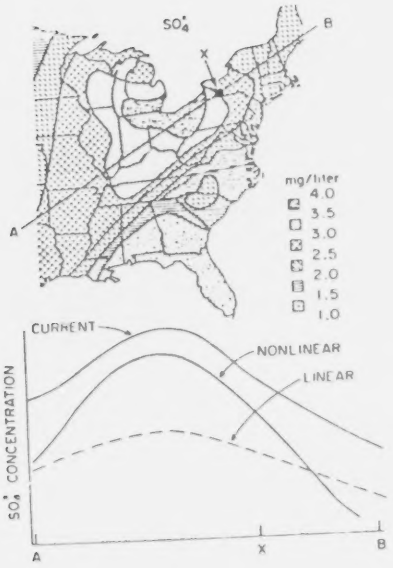


Fig. 1. Top--Observed 1979 average concentration of $SO_4^=$ in precipitation (after Wilson and Mohnen, 1982). Bottom--Current concentration along cross section A-B and hypothetical concentrations that would result from an emission reduction if the response were linear or nonlinear.

Reviewer D

CRITIQUE OF CHAPTER 4 —
EMPIRICAL OBSERVATIONS AND SOURCE-RECEPTOR RELATIONSHIPS

Introduction

Part of the thrust of the NAS report is that, given the present state of source-receptor modelling, we must rely on empirical observations to infer relationships between emissions and deposition and then to assess responses to mitigating strategies. One of the data sets presented for this purpose was the 16-year record of weekly wet deposition at the Hubbard Brook Experimental Forest in Central New Hampshire. While the emphasis on experimental observations may be appropriate at this time, the analysis presented was superficial and inappropriate to not only the Academy but to the importance and economics of the issue at hand.

The Hubbard Brook data have been extensively discussed and analyzed elsewhere.[1,2] But a serious attempt at a time series analysis relating these measurements to historical emissions of acid-forming precursors has not been presented, nor have quantitative relationships between wet deposition and air quality been assessed. Such a study is in progress and preliminary results are presented below.

The Academy Analysis (pp. 116-126)

The Academy analysis is based on a loose comparison of time trends between 1965-66 and 1979-80* for sulfate concentratons and emissions trends between 1965 and 1978 for any of several geographic regions. The emissions data are from the U.S. Canadian Work Group 3-B report,[3] which also reports data for two intermediate years, 1970 and 1975 (unfortunately, 1975 is a recession year and does not provide a reliable basis for trends between 1970 and 1978). Concentrations of nitrate in rain vis-a-vis NO_x emissions are also discussed, but more briefly. No statistical relationships are attempted between rain chemistry and emissions, nor are any confidence limits given for possible associations.

The trend in sulfate concentration (but not nitrate) is deduced from a linear regression analysis on 15 annual volume-weighted means, presumably giving each year equal weight. No explanation is given for excluding the two end periods, 1964-65 and 1979-80 (including them reduces the slope by about 10%). However, the trend in emissions over an equivalent period is deduced from only the two end points of the series, 1965 and 1978. The detailed behavior over the included time period is totally ignored as if the trend were monotonic (and linear) between these end points. Figure 4.15 shows that is decidedly not the case, even using the unrepresentatively low year of 1975 as a valid trend point. The fact is that 1970 (or thereabouts) is generally the peak year for SO_x emissions, after which the impact of the Clean Air Act of 1970 began to be felt. However, these reductions were realized slowly, so

*Hubbard Brook publications use a "water-year," beginning in June.

late all folios

that the years 1968-1974 are practically flat for national SO_2 emissions. The decrease began sooner in the Northeast, about 1972. In contrast, Figure 4.14 shows an increase in $SO_4^=$ concentrations during this period. To be fair, however, it should be mentioned that a linear regression of the individual weekly samples shows an overall drop of about 20% over 15 years (discussed more extensively below).

In addition to using disparate analytical methods for the concentration trends vs the emissions trends, the Academy report does not deal with the question of estimating which geographic regions' emissions have the most direct impact at Hubbard Brook. By implication, the analysis points towards the nearby locations (Federal Regions I and II) and would totally exonerate Region IV, where emissions increased 33% from 1965 to 1978.

A similar process is followed for nitrate vis-a-vis NO_x emissions. Including the two end periods in a linear regression on time increases the change from +8% to +23% (increase with time). The emissions increase, calculated from only the two end points 1965 and 1978, was 8.6% for Federal Regions I and II, whereas the NO_x emissions increase for Regions I-V was 27%. Thus, the implication as to whether NO_x has an influence from areas further away than New Jersey is critically dependent on which measurement years are used at Hubbard Brook. Using all the data to determine the trend would imply that more distant source regions should be considered for NO_x (but not for SO_2), an argument that runs contrary to the discussion on sulfate/nitrate ratios in the Academy report.

The Academy report concludes, with regard to the Hubbard Brook data, that there is "no evidence for a strongly nonlinear relationship between annual depositions and annual emissions in the Northeast." As a result, elsewhere in the report, the finding is reached that a reduction in emissions would result in a proportional reduction in deposition. This implies a 1:1 relationship or an elasticity of 1.0 [a 1% change in emissions would yield a 1% change in deposition (or concentration)]. It should be clear from the data in the Academy report that this finding is not supported, even with the limited analysis presented.

For sulfate concentrations, the following elasticities result from the Academy analysis, depending on the region of SO_2 emissions to be controlled:

Region I	1.15
Regions I and II	1.18
Regions I and II and III	0.67
Regions I - V	0.25

Thus, given the uncertainties of the analysis, the linear (i.e., proportional) hypothesis is supported if only the emissions from Regions I and II are considered for control, but not if Regions further south and west are included. The bulk of the report (and most of the extant literature on source-receptor relationships) makes it clear that such more distant regions should in fact be considered.

To illustrate the relationship between elasticities (as deduced from linear regressions) and hypothetical non-linear relationships, consider Figure 1:

Figure 1(a) shows three linear relationships with differing elasticities. The proportional relationship, $\epsilon = 1.0$, yields a 50% reduction in concentration (or deposition) for a 50% change in economics. However, if $\epsilon = 0.5$, only half the benefit is realized due to the presence of a y-intercept which could represent deposition arising from natural background or from sources outside the region of consideration. Converse arguments hold for $\epsilon = 2.0$.

Figure 1(b) shows how an actual non-linear relationship ($C = \sqrt{E}$) might be represented by linear approximation, for example derived from regression analysis. A linear regression through the entire curve (11 points spaced every 10% in emissions) yields a correlation coefficient of about 0.96, explaining 92% of the variance (which would be considered an excellent fit). The slope (and hence elasticity of this line is 0.87, which is not very different from unity. Yet if reductions in emissions are postulated of the order of 50%, the asymptotic linear approximation shows that only about half of the expected benefit would be realized in fact. It should be noted that in most cases, we are working with the upper ends of the curves in question (the range in National SO_x emissions from 1965 to 1979 is about 5 million tons out of a total of about 30 million.

It should be clear from this example that it is extremely unlikely that statistical analysis of time trends will be able to detect nonlinearities per se. However, differences in the elasticities of linear best-fit relationships should indicate the degree of proportionality to be expected. The range of elasticities given above, based on the figures in the Academy report, indicate that no conclusions on linearity or proportionality can be made from this analysis.

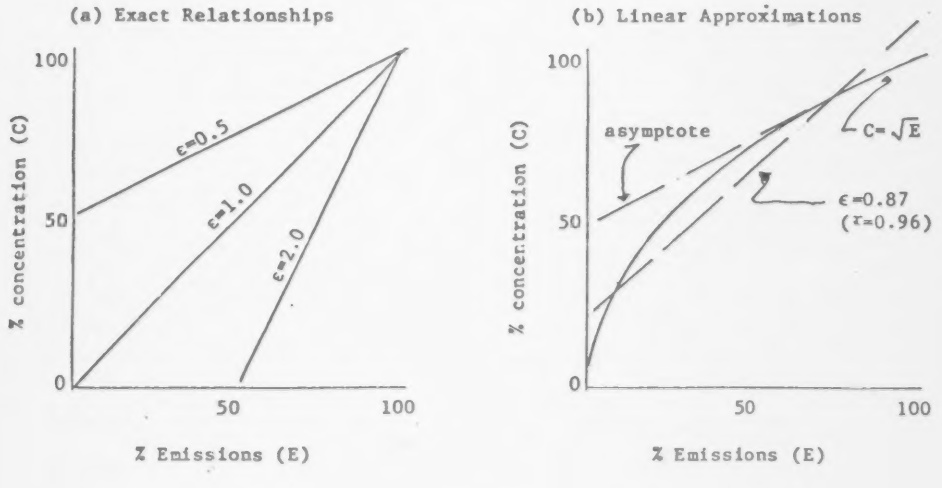


Figure 1

Reanalysis of the Hubbard Brook Precipitation Chemistry Data (1964-79)

Because of the importance of this data set, a reanalysis of the weekly (bulk) samples has been carried out, using multiple linear regression methods. The dependent variables were the amounts of precipitation in each weekly sample and concentrations (μeq/ℓ) of the following ions: H^+, $SO_4^=$, NO_3^-, Cl^-, Al^{+++}, NH_4^+, Ca^{++}, Mg^{++}, K^+, SiO_2, $PO_4^{\equiv}$ and Na^+. Concentrations were corrected for sea salt content assuming that all the sodium is derived from that source. Further, the balance between positive and negative ions was calculated, and only the observations for which this balance was achieved within ±20% were retained (about 87% of the observations or about 440 values).

The following independent meteorological variables were used:

PRECIP — the amount of each weekly sample

WAVE — a periodic function intended to be a surrogate for such seasonal effects as photoperiod, wind trajectory variations, fraction of convective vs continuous storm, etc.

$$\text{WAVE} = \cos\left[\left(\frac{\text{day of year}}{365} \times 360\right) - 180 \ * \frac{\pi}{180}\right]$$

MAVTEMP — monthly average surface temperature at Concord, NH (finer time resolution not readily available).

The influence of trajectory differences by year at Hubbard Brook was considered in detail by Munn et al.[2] SO_4 concentrations were normalized by accounting for the frequency of occurrence of 5 different wind direction sectors; concentrations tended to be higher for trajectories originating to the south and west of Hubbard Brook than from the east, for example. However, this normalization had only a trivial effect on the actual annual median SO_4 concentrations, increasing the overall mean about 1-3%, but with virtually no change in the standard deviation from year-to-year. Thus, by implication, the differences shown from year-to-year in median sulfate concentration are not due to trajectory variations. The assumption that this source of variation may be ignored in a multiple regression analysis without creating bias appears to be well-founded.

The emissions data were computed for each year from 1964 to 1979 based primarily on fuel use data[4] at the state level for the United States. Statewide fuel sulfur contents were derived from FPC Form 67 data from 1969 to 1976. For years prior to 1969, these values were assumed constant. For years after 1976, emissions by state were estimated from published EPA reports. In general, this approach captures only combustion-derived emissions, a limitation that is not believed to be serious for the Eastern United States. Canadian copper-nickel smelter emissions[5] were also included for those years, as was an estimate of other eastern Canadian emissions,[6] for which only 2 years' data were available requiring linear interpolation to represent the 16 year period.

Quarterly shares of emissions were available for 2 years (1965 and 1978),[9] for the SURE region (Northeastern U.S.). The differences between these shares and a uniform annual distribution were minor, with higher emissions in the winter. When used, these quarterly shares were applied to all emissions estimates, regardless of geographic region. A conservative estimate of the number of degrees of freedom for the emissions data is then 18 (16 annual values plus 2 quarterly shares). Comparison of these independent emissions estimates with those of EPA shows somewhat higher estimates but similar trends (for the years available from EPA).

As an alternative to aggregation by summation, weighting schemes were considered in order to combine emissions from the various geographic regions thought to have an influence at Hubbard Brook. Both inverse linear and inverse squared weightings were used, based on distances from Hubbard Brook to the estimated emissions centroids of the regions.

Multiple linear regressions were performed, using the stepwise mode and the meteorological variables listed above together with various emissions variables. The regression step selected for detailed consideration was either the one with the lowest standard error of estimate (highest adjusted R^2) or the subsequent step upon which the emissions variables entered the regression. Examination of residuals did not indicate autocorrelation to be significant, based on the Durbin-Watson statistic.

It was hypothesized that sulfate deposition at Hubbard Brook derived from two separate sources: aerosol which had been scavenged by precipitation, and sulfate formed in clouds. The aerosol portion was proportional to the concentration of ammonium ion (NH_4^+) in precipitation. Thus, the balance ($SO_{4eq}^{=} - NH_{4eq}^{+}$) would be a surrogate for the sulfate formed in clouds. An alternative estimate of in-cloud sulfate subtracted the calcium equivalent in addition ($SO_{4eq}^{=} - NH_{4eq}^{+} - Ca_{eq}^{++}$), recognizing that calcium is an important ion at Hubbard Brook, with a substantial downward time trend. However, since calcium is thought to be derived from suspended dust of coarser particle size that ammonium (sulfate), these two ions were not summed to represent aerosol derived from anthropogenic emission sources.

Table 1
Elasticities and 95% Compliance Limits

Relationship	Emissions Region	Summed Emissions	Linear Weighting	Squared Weighting
NO_3^- vs NO_x	State of NH	0.75±0.36	--	--
	Region I	0.25±0.45	--	--
	Regions I - II	0.70±0.85	0.89±0.62	0.70±0.51
	Regions I - III	1.41±0.58	1.17±0.69	0.84±0.56
	Eastern North America	1.12±0.61	1.03±0.57	0.88±0.51
	All U.S. + E. Canada		0.88±0.47	0.84±0.47
$SO_4^=$ vs SO_2	State of NH	0.05±0.30	--	--
	Region I	-0.27±0.44	--	--
	Regions I - II	-0.59±0.62		
	Regions I - III	-0.42±0.84		
	Eastern North America	-0.22±0.88		
	All U.S. + E. Canada		0.59±0.71	±0.42±0.46
NH_4^+ vs SO_2	State of NH	0.60±0.52	--	--
	Region I	0.47±0.35	--	--
	Regions I - II	0.55±0.43	0.53±0.40	0.51±0.38
	Regions I - III	1.01±0.68	0.81±0.56	0.63±0.46
	Eastern North America	1.00±0.67	0.96±0.65	0.81±0.57
	All U.S. + E. Canada		1.94±1.19	1.18±0.77
($SO_4^=$ - NH_4^+) vs SO_2	State of NH	-0.11±0.33	--	--
	Region I	-0.49±0.48	--	--
	Regions I - II	-0.91±0.68	0.11±0.26	0.10±0.24
	Regions I - III	-0.93±0.92	0.15±0.36	0.12±0.29
	Eastern North America	-0.73±0.96	0.21±0.42	0.17±0.37
	All U.S. + E. Canada		0.20±0.77	0.20±0.50

Table 1 presents preliminary results from this analysis, based on the following:

- Only data for which an ion balance was achieved within ±20% were retained.
- Annual rather than quarterly emissions were used (elasticities are slightly lower and less statistically significant when the quarterly values are used).
- 95% confidence limits are based on 15 degrees of freedom (t = 2.13).
- Emissions are aggregated by Federal region in three different ways: summation, weighting by the inverse distance from Hubbard Brook to the estimated centroid of emission, and weighting by the square of that distance.

Nitrate Ion

Statistically significant relationships were shown for almost all cases. In most cases, the elasticities are not significantly different from unity. There is no reliable indication as to which emission region provides the best fit, but there does seem to be a tendency towards an area larger than Federal Regions I and II.

Sulfaction

There are no statistically significant relationships between sulfate in rain at Hubbard Brook and SO_2 emissions for any of the regions considered. However, the weighted "all U.S. + Canada" cases nearly achieved significance with an elasticity around 0.5 (most likely value).

Ammonium Ion

Ammonium ion (taken here as a surrogate for sulfate derived from aerosol) shows statistically significant relationships with SO_2 emissions in all cases. Moreover, as the region increases in size, the elasticity increases, indicating the need to control over a wide region for maximum effectiveness. However, as with nitrate ion, it is not possible to indicate the optimum region for control with any degree of confidence.

Sulfate less Ammonium

The difference between sulfate and ammonium equivalents is interpreted here as a surrogate for sulfate formed in clouds. No positive statistically significant relationships are seen. Since

it is difficult to conceive of a physically meaningful negative relationship, the weighted results appear to be the most credible and indicate no relationship between the two variables. This could indicate a situation of saturation, in which no more sulfate is formed if fresh SO_2 is supplied, and no less if SO_2 is reduced. Such a situation can only pertain locally, due to global mass balance considerations. Subtracting calcium in addition to ammonium ion did not materially change these results.

Figures 2 to 5 present plots of "seasonally adjusted" annual mean concentrations vs emissions. The "seasonal adjustment" procedure accounts for the effects of precipitation amount, monthly average temperature, and the seasonal periodic function (WAVE) by subtracting these effects as predicted by the regression equation, according to the following formula:

$$C_{seas.adj.} = C_{measured} - (K_1 * PRECIP + K_2 * WAVE + K_3 * MAVTEMP)$$

where the K's are given by the averages of coefficients for several regression equations.

Figure 2 presents the time trends for sulfate in precipitation at Hubbard Brook and for the linearly (inverse) weighted SO_2 emission parameter, which includes all of the U.S. and Eastern Canada. After 1970, the trends are reasonably concordant, but before 1970 they are opposing. The $SO_4^=$ plot looks much like Fig. 4.14 in the Academy report, even though weighting by precipitation volume has not been done in Figs. 2-5.

Figure 3 presents the cross plot, which indicates a weak (non-significant) positive relationship between the two parameters, with an elasticity of 0.35. An elasticity 1.0 line is also shown for comparison, and the two years used in the Academy report (1965 and 1978) are flagged. Note the very different impression created by considering just these two years.

Figure 4 presents a similar plot for ammonium ion, but in sharp contrast the data fit an elasticity $\simeq$ 1.0 line with a highly significant correlation ($P < 1\%$). Figure 5 presents nitrate data vs the linearly weighted (inverse) NO_x emissions parameter, which also results in a highly significant correlation, but a lower elasticity than obtained with the weekly sample analysis. The use of only 1965 and 1978 would provide a reasonable estimate in this case.

Conclusion

This analysis has indicated that at Hubbard Brook, proportional reduction might be expected in nitrate and ammonium deposition from the control of NO_x and SO_2 emissions, respectively. Considerably smaller benefits if any, would be expected for deposition of sulfate ion.

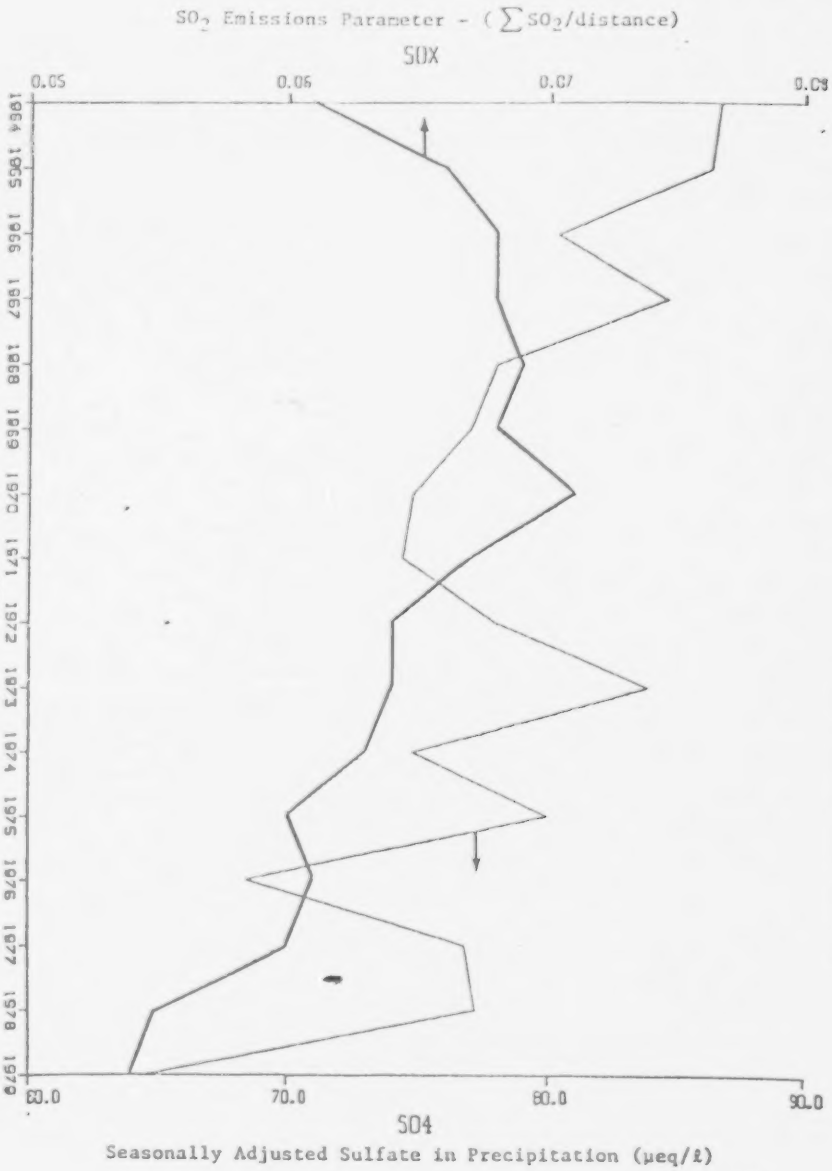


Figure 2. Time Trends for SO_2 Emissions and Precipitation Sulfate

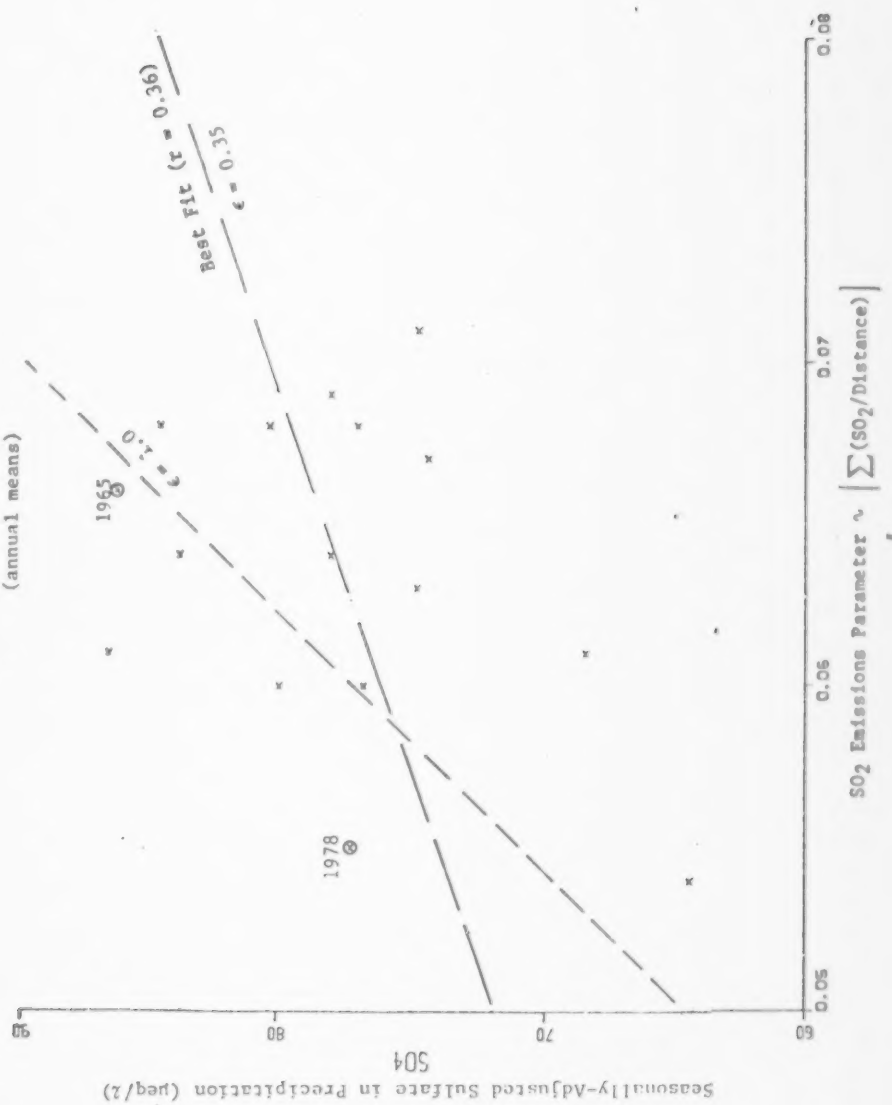


Figure 3. $SO_4^=$ vs SO_2 Emissions
(annual means)

Figure 4. NH_4^+ vs SO_2 Emissions
(annual means)

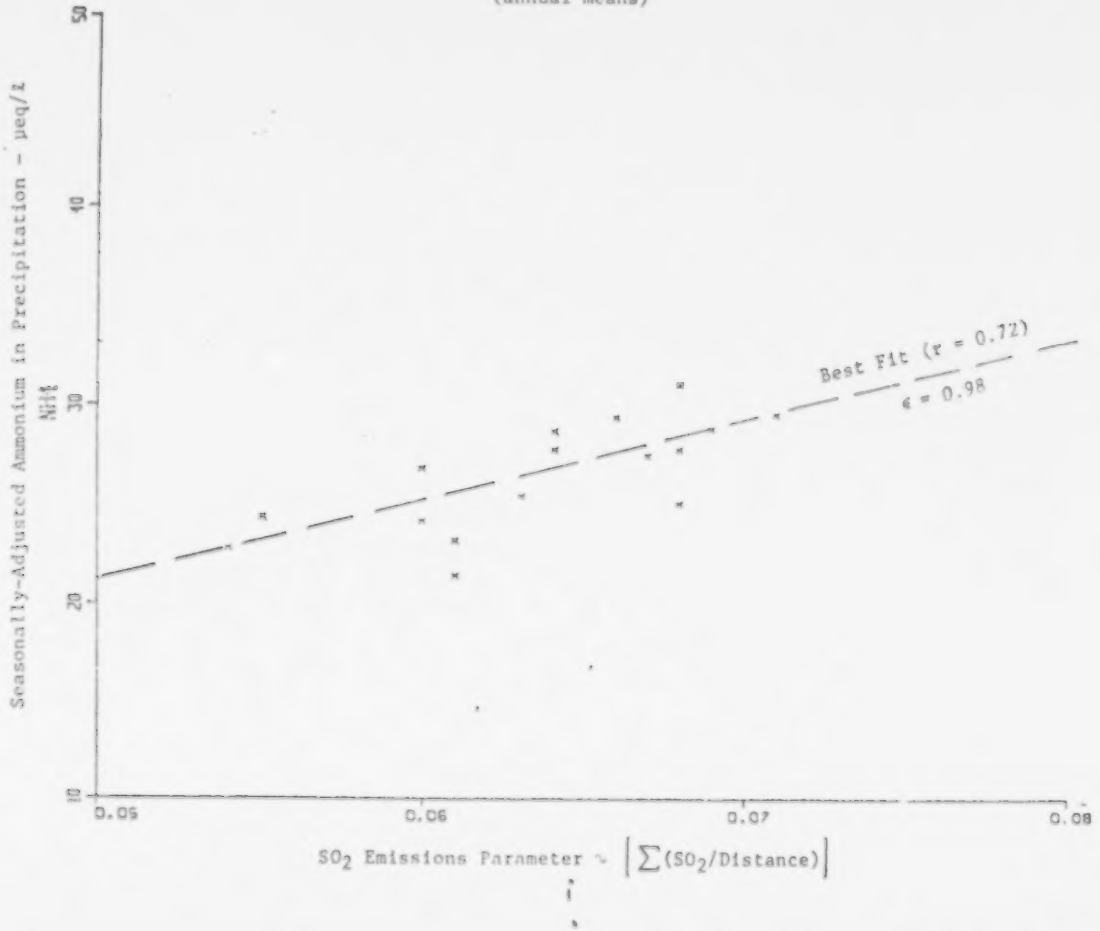
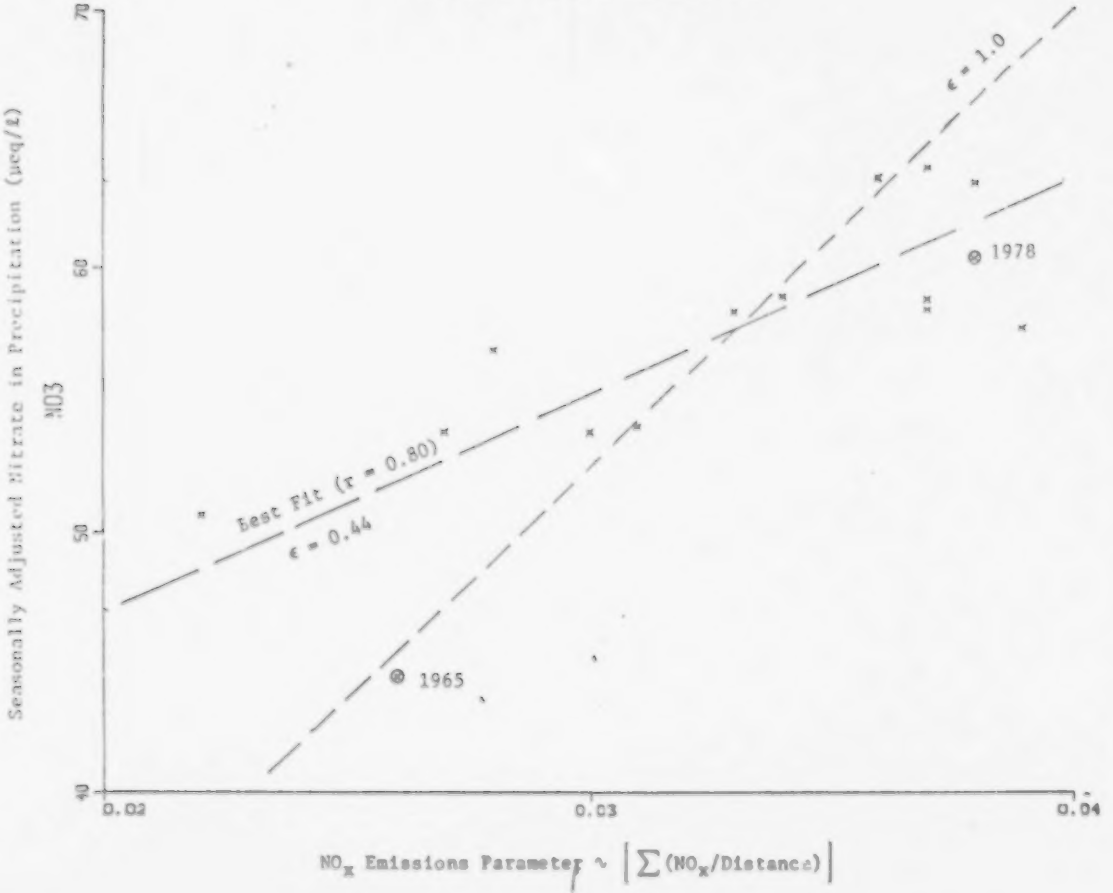


Figure 5. NO_3^- vs NO_x Emissions



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Reviewer E

Review of the report by the National Research Council (Environmental Studies Board of the Commission on Physical Sciences, Mathematics, and Resources)

"ACID DEPOSITION -- ATMOSPHERIC PROCESSES IN EASTERN NORTH AMERICA -- A Review of Current Scientific Understanding",

which was prepared by the Committee on Atmospheric Transport and Chemical Transformation in Acid Precipitation, with membership: J. Calvert (Chairman), J.N. Galloway, J.M. Hales, G.M. Hidy, J. Jacobson, A. Lazrus, J. Miller, and V. Mohnen.

(This review was written (during a rushed, 40-hour weekend) by Reviewer E , who has published approximately 50 open-literature papers in the fields of precipitation scavenging and dry deposition during his past 16 years of research in these fields)

INTRODUCTION TO THIS REVIEW

This report published by the NRC is very strange. If one were to start reading at Chapter 1 (rather than first read the cover letter by F. Press and the Summary), then the first half of the book (and the associated appendices) flow along quite well. Oh, there are small problems, but nothing too serious. Thus, on p.24, L9, the goals of the project are clearly stated:
"If this goal (reducing the hydrogen ion deposition in sensitive regions by 50 percent) were adopted, by how much would emissions have to be reduced? Conversely, by how much would deposition rates be reduced if there were specific reduction in emission rates?", and on p.24, middle:
"Our objective was to determine what conclusions can be drawn from the state of knowledge late in 1982 about the relationships between emissions and deposition."
Some small problems encountered include the weak description on pp. 30-35 of transport and mixing (even when Appendix B is added), the tendency to jump on the "H2O2 oxidation bandwagon" (what a lot of red faces there will be -- or should be -- if HNO3 is found to be the dominant aqueous-phase oxidizer of SO2), weak descriptions of wet and dry deposition (but well compensated by Appendix C), and an error in describing the influence of gradients of diffusion at the bottom of p.50 (see later).

But then things start going more poorly in Chapter 3. After first reading the description of models (pp. 55-72), I looked again to see who were the authors. As expected from the poor description of the models, I found that not one of the authors is a long-range transport modeler: the committee was overwhelmingly dominated by chemists and chemical engineers.

Then, halfway through the book, the excrement definitely becomes entangled with the impeller. On p. 69, middle, we gain the first hint that the Committee does not plan just to review current work that is available to the scientific community, but instead plans to plow into a few virgin fields by themselves.
"It is almost certainly true that the scheme of Rodhe et al. fails to do this (include the essential chemistry) properly."

That's all it says. No reference. As yet, no alternative. Just: Rodhe et al. are wrong. <I'll come back to this point later, but let me at least mention the essence here. It seems that the chairman of the Committee has recently coauthored a paper -- accepted for publication but not yet available for study by the scientific community -- that purports to have a better homogeneous gas-phase oxidation scheme for SO2 than the one used by Rodhe et al. Yes, gas phase; yes, gas-phase oxidation of SO2 may be ignorable; yes, the Chairman's.

And then, on p.72, it happens. They stumble into it this way (3rd paragraph):

"Recently, in connection with proposals for mitigating acid deposition, the question has arisen as to whether deposition rates are linear functions of emissions."

Here we are, halfway through the book (or farther, if account is taken of the appendices), and this is the first time the linearity/nonlinearty issue arises. THE COMMITTEE THEN BECOMES ENGROSSED IN THIS ISSUE FOR THE REST OF THE BOOK, ROYALLY SCREWS IT UP, PRESENTS UNSCIENTIFIC, SUBJECTIVE, HAND-WAVING, ATROCIOUS "ARGUMENTS", ABANDONS ANY LINK WITH CURRENT SCIENTIFIC OPINION, AND DISREGARDS THE STATED OBJECTIVES OF THEIR STUDY. And it is this linear/nonlinear issue that is picked up by Press and the press.

Worst, as chemists, they focus on chemical nonlinearity (e.g., not first-order reactions) and thereby, almost totally disregard the heart of the nonlinearity issue (I say "almost", because this idea can be found in odd sentences in the text, but not in the summaries). The heart of the nonlinear issue is not whether the reactions are linear in concentrations, but whether there are any reactants available to react. That is, at heart, the question is: what is the spatial dependence of the deposition? This the Committee ignores, confining their attention in the rest of the book to large-scale spatial averages. The second-half of the book flows steadily downhill, save for the well-done chapter on research needs (if the reader can tolerate the Committee's use of the English language -- I could not).

In view of the disasterous second half of the book and in view of the emphasis on the second half that is contained in Press's cover letter and in the summary, I propose here to start this review at the beginning of the book (with the Cover Letter and the Summary), but give references to where in the text the summary material is coming from. Then, if I have time left during this working weekend, I'll try to type out my review of the text. My apologies to readers (if there are any) for typing, spelling, and grammatical errors. I do not have the time to review this production, and my Apple and I are just becoming acquainted (and words seem to be spelled and arranged differently on this screen from their appearance on paper!).

THE REVIEW

I. Title Page (p. i):
1. It should be "Acidic Deposition", not "Acid Deposition"
2. It will be seen that there is very little in this report on "Atmospheric Processes"; the authors are overwhelmingly chemists, not meteorologists.
3. And it will be seen that this should not be called a "Review of Current Scientific Understanding". A more appropriate subtitle would be: "Some Wild Speculations about Meteorological Processes by Chemists".
4. I am not impressed by the list of "responsible" groups (Committee..., Board..... Commission.....Council); in reality, the speculations in this book are the responsibility of eight people, mostly chemists.

II. NOTICE (p. ii):
1. L.6: "The members of the committee <sic> responsible for the report were chosen <by whom?> for their special competence <in what?> and with regard for appropriate balance." <of what?>
Stating something doesn't make it so. There is an overwhelming number of chemists on the committee, none of the Committee members has experience in modeling long-range transport and deposition of air pollution, and there are no representatives of the industries that would be influenced by the wild speculations that appear in this report.
2. L.9: "This report has been reviewed by a group other than the authors".
I would hope so! -- and I also would have hoped that the other group would have been more knowledgeable and less imbalanced by chemists, but apparently that was not the case.
3. L.23: "The Council has become the principal operating agency ... in the conduct of their services to the government, the public, and the scientific and engineering communities."
Most service will be provided by sticking to science. As scientists, we certainly must engage in wild speculations (also known as hypotheses). But these speculations/hypotheses are then to be tested, not published under a title of "A Review of Current Scientific Understanding."

III. Cover letter by F. Press, Chairman of the National Research Council (p.iii):
1. P1: "acid" vs. "acidic"
2. P2, S2: "The public policy implications of this relationship <of linearity vs. nonlinearity> are of major economic and political significance."
That is true, and it is therefore even more important that wild speculations be avoided -- but they are not.
3. P3, S2: "The Committee concludes that there is no evidence that the relationship between emissions and deposition in northeastern North America is substantially non-linear when averaged over a period of a year and over dimensions of the order of a million square kilometers."
I will return to this conclusion. Here, I only want to state

that the Committee also gives no evidence that the relationship is linear. Moreover, overwhelmed by chemists, the Committee doesn't seem to understand even the meaning of "linear" vs. "nonlinear". The Committee does, however, give evidence (but chooses to ignore it) that the system is saturated. Therefore

4. S3: "The Committee's judgement that if the emissions of sulfur dioxide from all sources in this region were reduced by the same fraction, the result would be a corresponding fractional reduction in deposition" does not conform with the evidence that the Committee gives, but chooses to ignore.

5. p.iv, P1 (if it is a paragraph! -- it lists the "several pieces of evidence", mentioned on the previous page), S1: "One is the well documented observation that the molar ratios of sulfates to nitrates deposited at many observation points in this region are nearly constant, and are similar to the molar ratios of the integrated emissions."

Of course I will respond to this point again, but even now, I would ask the reader to turn to Figs. 4.17 and 4.18 (pp. 130 & 131). This is the "well documented observation" that is being referenced. Look first at Fig. 4.18, showing the molar ratios of the emissions. I defy anyone to try to supply contours of constant ratios on this figure -- it looks like the results from a random number generator. Now look at Fig. 4.17 (and it would have been nice if the Committee would have included the data for Newfoundland). But look what the Committee did add: contours! A pattern! Giving what? Increasing sulfate to nitrate, farther from the source. What does this suggest? That the system is saturated! Moreover, EVEN IF THE MOLAR RATIOS WERE CONSTANT (BUT THEY'RE NOT!) THEN THE SYSTEM COULD BE SATURATED: if there is enough oxidant available to oxidized only a fraction of either NOx or SO2, then we don't KNOW if NOx would be preferentially oxidized IN PRECIPITATING SYSTEMS. In other words, this "well documented observation" doesn't suggest what the Committee says it does (in reality, the molar ratios show a definite trend, increasing farther from the source), and even if the ratio were a constant, that doesn't mean that the system is linear -- it would only mean that the ratio is constant, which could be explained in many ways.

6. S2: "The Committee believes <sic> that the simplest and most direct interpretation of this observation is the one they have given."

I agree: the Committee's interpretation certainly is simple minded. And I think that the word "belief" is highly appropriate. "Lief" is latin for wish; "belief", therefore, means "wish to be". In science, however, we must dispense with "liefing": when a scientist starts "wishing" for something to be (other than a belief in objectivity), then science becomes dogma. Who cares what reality is; let's just describe it, objectively.

7. S3: "A second observation that supports the Committee's judgment comes form a 13-year run of observations at a single location in North America that shows the linear relationship between deposition at this one site and overall emissions."....and the price of rice in China.

Would the reader now please turn to Fig. 4.19, p.134. The data

points scattered there (S4) "represent the result of well designed and carefully taken measurements" -- from a bucket left open for a full week, exposed to local dirt, leaves, bugs, bird droppings, near a power plant that was shut down in 1970 (note the decrease in sulfate in 1970), and near a highway on which traffic decreased in the early 70's (decreasing nitrates, and therfore increasing the ratios). No, I don't know if that is true -- but there is nothing in the report to justify the statement "well designed and carefully taken measurements", and again and again the authors of the report state that they can't evaluate the contributions from distant vs. local sources. In other words, the Committee repeatedly states that they can't trust single-point measurements, but here we are told that these single-point data form one of the "several" (three!) "pieces of evidence" on which their "judgement is based."

And what, I hope the reader will ask, are the other curves in Fig. 4.19? These curves are supposed to represent the molar ratios of emissions in the various EPA Regions (Regions I and II include New England and New York). Guilt by association? But I think that, in reality, the curves represent the confidence in the National Academy expressed by five scientists: steadily decreasing with time. How dare the National Academies put up a set of curves, give no scientific (hypothesis/test) basis for a correlation, yet suggest a correlation, and also suggest that this is "in the conduct of their services to the government, the public, and the scientific and engineering communities." Thank you, but if this is science, I withdraw my membership in the scientific community.

8. End P1: "Finally, the judgment draws upon the results of recent laboratory studies that are discussed in the report." Now of course it's difficult to pin them down on where they are "discussed in the report", but it appears that the major studies are those mentioned in the "Findings and Conclusions" on pp. 83 - 84 (top paragraph on p. 84: "On the basis of laboratory evidence, we conclude... Laboratory evidence suggests ..."). In turn, these studies are described on p. 78 - 79, where three points are blatantly apparent: (i) The referenced paper is not yet published and therefore has not been evaluated by the scientific community (which typically takes from 5 to 50 years), (ii) The referenced paper deals with homogeneous, gas-phase chemistry (and therefore is likely almost irrelevant to the issue of acidic rain), and (iii) The paper happens to be coauthored by the Chairman of the Committee.

And those three sum to give the evidence from which billion-dollar decisions are to be made? Joy!

9. P2, S1: "The Academies' process of selecting a diversified, expert study Committee and subjecting the Commitee's report to a substantive review was carefully followed in the preparation of this document."
Has anyone tested the process to see if it works?

10. P3, S1: "I believe..." Yes, I can understand that.

IV. Preface by J. Calvert, Chairman of the Committee. Bottom of p.ix, top p.x: It is important, I think, to take note of what was asked of the Committee (for later comparison with what the Committee produced). "... the Chairman of the NRC asked the Environmental Studies Board to review the current state of knowledge about atmospheric processes that link emissions to deposition" -- this was not done: the review of knowledge about long-range transport and models (e.g., Appendix B) is woefully inadequate -- "with the purpose of describing to the extent possible the consequences of the goal of reduced deposition for emissions." <sic -- whatever those words mean!> "By how much would emissions have to be reduced to reduce the deposition of hydrogen ions by 50 percent in sensitive areas? Conversely, by how much would depositions be reduced if there were specific reductions is emissions?"

And how great it would have been if the Committee could have seen behind the veneer and had the courage to describe what they saw. Oh, they have tried to help: but this way, in the long run, they do more damage than good. Can you imagine their saying: "Look, people, let's be realistic. Our society chose to release these huge quantities of SO2 and NOx to the atmosphere, and decided not to examine the consequences. Thereby, many of our scientific colleagues are unemployed, or driving taxis, selling real estate, farming, etc. These are honorable jobs, of course, but the parents of these scientists worked to realize their hopes, and the students worked very hard to learn science. If they had known that there was no employment in science, they would have skipped the effort, and gone directly from high school to these jobs, or trained for other jobs. And now, Mr. and Ms. Public and your representatives, you come to us and ask what to do about the pollution. Well, we're sorry, but we don't know. You never funded us to find out. Sure we could have told you, and we can yet, if you are prepared to fund us. But no, we're not going to engage in wild speculations about what to do about the pollution. That way we weaken science. If we did supply you with some wild ideas, any scientist worth the name will stand up and say: "That's bunk!". What then? We would have weakened science. And in the long run, Mr. and Ms. Public, that will hurt you more than the pollution will."

V. Summary
1. p.2, last line: What is/are observational data? Is there some other kind?
2. p.3, L.9: "A long-term record (18 years)" <I thought Press said 13 years> "of reasonably reliable data" <How reliable is "reasonably reliable"? Press said "they represent the result of well designed and carefully taken measurements">.
3. p.3, end of P.2: "Currently the molar concentrations of nitrate and sulfate in precipitation are roughly comparable over much of the eastern United States." How rough is "roughly comparable"? Why did Press say "northeastern North America" and here it says "eastern United States"?
4. Para at bottom of p.3 top of p.4:
Here's a little game. Take the following three ingredients
(i) "Concentrations of sulfate in both the air and precipitation tend to reach their maximum values in summer in the northeastern United States"
(ii) "Nitrate in precipitation shows much less seasonal variation in the northeastern United States"
(iii) "Currently the molar concentrations of nitrate and sulfate in precipitation are roughly comparable over much of the eastern United States".
The game is to determine how those three statements can be consistent. <The answer: bury it all in the word "roughly"! It's a great game. It can be played on and on: all concentrations are "roughly" at background levels; the system is "roughly" linear; "roughly", none of the pollution stresses the ecosystem;... Joy!>
5. p. 5, L9: What, pray tell, do the authors mean by "empirical analyses". My Webster gives me two options for the word empirical: (i) relying or based solely on experiment and observation rather than theory, or (ii) relying or based on practical experience without reference to scientific principles. If the authors are using "empirical" in the second sense, then I presume that the analyses are based on when the squirrels store their nuts, as recorded in the Farmer's Almanac. If the authors are using "empirical" in the first sense, then how can the adjective empirical be used to modify the noun analyses; it's like saying "nontheoretical theory". In either case it's sensible nonsense.
6. p.5, end of top Para: "But the relative contributions of specific source regions to specific receptor sites currently remains unknown."
What is meant by "unknown"? Is it known to within a factor of 2? Factor of 10? If it's known to within 27 decimal places, it can still be said to be unknown! The quoted statement is totally useless -- except as garbage words to fill up the page, as with "observational data" and "empirical analyses". I know these relative contributions to within a factor of 5, with 95 percent confidence; so how is it that the authors find that the "relative contributions ... currently remain unknown"?
7. Suppose I just ignore the "theoretical calculations". Maybe it is the case here, as with most chemists, that any calculation they perform is to be treated as an experiment -- with uncertain outcome!

8. But I can't let pass (p.5, P2, S3): "The methods employed to date, however, do not produce reliable estimates of spatial and temporal distributions of acidity..." Isn't that a beautiful scientific statement? Isn't it great to belong to such a profession?
"Doc, tell me, what's the chance that this operation is gonna kill me."
"Well based on observational data and empirical analyses, the answer is currently unknown."
"Ya sure Doc, but am I gonna live."
"Well, the methods employed to date do not produce reliable estimates."
"Gimme a number!"
"The actual limitations of predictive models ... are not well defined because of deficiencies in knowledge..."
"See ya around Doc."
9. p.6, L18: "However, current models have not provided results that enable us to have confidence in their ability .."
And statements such as that don't engender much confidence in the scientific abilities of the writers. How much "confidence"? What are the confidence limits? In the next paragraph: "Workers in the field generally have only limited confidence in current results." -- Be specific! A few more wishy-washy statements like that and this whole report will turn into slop.
10. p.7, middle para: Where did the laboratory studies all of a sudden come in? Why the comment about gas-phase reactions? Who decided that gas-phase oxidatation of SO2 was so important as to be included here, under "ISSUES"? The chairman?
11. p.7, bottom: I've already commented some on this nonlinearity issue, and I'll return to it again. For now, I'll let it pass.
12. p.8, L7: "The only available direct evidence of strong nonlinearity between average emissions and average deposition of sulfur compounds is found in the extensive data taken in Europe over the past 25 years."
Why the word "only"? Or if only is to be used, how about only using it to describe the data as the only useful data available? In my opinion, the only scientifically satisfactory statement that the committee could now make is: "This is the only extensive data base available anywhere in the world, and though hypotheses can be made that there would be differences in North America, until data are available that test these hypotheses, we can only call the reader's attention to the European result."
13. p.8, middle: "These data (from the Hubbard Brook site) show no evidence of strong nonlinearity."
This is the kind of statement that makes me sick. The authors are scientists? WHAT, PRAY TELL, WAS YOUR STATISTICAL TEST? (And for those who can't stomach reading this whole book, let me tell you the answer: NO TEST! They drew 5 or 6 lines on a graph paper and said: "Hey look, they're all goin' the same way. Gee, there mustn't be any nonlinearity here." -- The lines? Does it matter? I think one was the height of women's hem lines, another was....)
14. p.8, 15 lines from the bottom. And now the report states "eastern North America", not just the U.S. What's it gonna be?

15. p.8, 3 lines from the bottom: "It is therefore improbable..." Please spare us from your weasel words. A few more "improbables" and "little confidence" and "advise caution" and I'm gonna get mad.
16. p.9, L3: Oh beautiful. Isn't that dandy: "The North American data therefore suggest ...." One plot that shows that the molar ratio DOES increase with distance (that's opposite from what the Committee says), and one time series from a single bucket, collecting local bugs -- these are THE North American data!? How about "from just two of the hundreds of sets of data that we could have examined, and from these two sets to which we have applied no objective, statistical tests, but instead, have used subjective eyeballing, learned in kindergarden, we guess that: 'Aw shucks, why not go ahead and spend the $100 billion and see what happens'."
17. p.9, middle: more of this "cannot be reliably estimated.." How I long for just one quantitative statement in this report.
18. p.9, second to last para: "Trends ... appear to reflect..." Joy!
19. p.9, 4L from bottom: "atmospheric processes ... lead to thorough mixing .. making it difficult to distinguish between effects of distant and local sources."
Isn't the information content in this report staggering?! Dear Editor: I have reviewed this report as you requested, and recommend that you through it in the trash.
20. p.10, top: Follow this logic if you can:
(i) "The implications of our findings and conclusions ... are limited." <Don't worry about what we're gonna say; the implications aren't important>
(ii) "It is <not> practical ... to rely on currently available models.." <So we won't do that>
(iii) "Data povides guidance" <So of course we're gonna use the data>
(iv) "If we assume ...."
HELLO. DO I SMELL A MODEL?
(v) "The annual average concentration of sulfate in precipitation at a given site should be reduced in proportion to a reduction in SO2 and sulfate transported to that site form a source or region of sources."
PRAY TELL WHAT DATA PROVIDED THIS GUIDANCE?
Oh, yes but of course, what's a few hundred billion more or less.. the "implications are limited"!
21. p.10, P3: But undaunted, this fountain of wisdom keep flowing: "It can be stated as a rule of thumb that the farther a source is from a given receptor site, the smaller its influence on that site will be per unit mass emitted."
Thank you. I wonder if we could survive without these chemists -- I sure as hell would like to give it a try.
22. p.10, next sentence. "Analysis of air-mass trajectories and modeling may provide insight..." Would someone please put a muzzle on these guys! "Interpretation of this information, however, is subjective .." If so, I nominate these chemists -- subjectivity seems to be their rule of thumb, and they're all thumbs.
23. p.10. The last paragraph is a gem. They give a totally

unsubstaniated, subjective opinion, and then state that they can't rely on models and therefore "cannot objectively predict the consequences ..." Don't get hung up on objectivity now, fellas; the lack of objectivity hasn't seemed to bother you up to now in this report.

24. p.11. S2 in last para: "It appears to us, however that useful information ... can be determined more quicily by direct empirical observation in the field.."

Beautiful!

"Observations", I understand.

"Direct observations", yes.

"Direct observations in the field"? How does one perform indirect observations in the field? Or direct observations of meteorological processes in a laboratory?

"Empirical observations"? Empirical in the sense of "relying or based on practical experience without reference to scientific principles"? What a strange recommendation coming from the National Academy of Science.

"Direct empirical observations"? What are INDIRECT empirical observations? If direct vs indirect, is there then some scientific principle? But that's not empiricism. Or do they use "empirical" in the sense of "relying or based solely on experiment and observation rather than theory"? Then what is an empirical observation? And why neglect the other half of science. Yes, SCIENCE. Oh, you people don't know about science? Hmmm. Yes, then, perhaps its best. Why don't you chemists just continue on with your "direct empirical observations in the field" -- far better you do that then engage in more of your wild speculations.

VI. Introduction
1. p.14, 7L from bottom: back to the "long-term (18-year) record of reasonably reliable data"
2. p.24, end of top para. The goals are stated clearly: "If this goal (reduce hydrogen ion deposition by about 50 percent) were adopted, by how much would emissions have to be reduced? Conversely, by how much would deposition rates be reduced if there weere specific reductions in emission rates?"
It's a pity that, when this draft (now, the final draft) of the report was finished, the authors didn't review their report to see that they didn't meet, or even come near meeting, these goals.

And they could have. For example, suppose they were to focus on deposition in the Hubbard Brook area. Available models of transport, diffusion, transformation, and deposition could predict the weekly-average deposition of sulfate and nitrate from sources within about 100 km, within known confidence limits. I expect that sensitivity analyses would show that the predictions were within a factor of two of reality. Then, available long-range transport models could precict the Hubbard Brooks' deposition from all sources, and the result could be compared with the data. I expect that sensitivity analysis would show that the comparisions with data were within a defined range of uncertainties of all parameters. Then, a different set of predictions could have been made and compared with an independent data set (e.g., at the MAP3S sites). After these exercises, I expect that the stated goals could have been satisfied for any region, with known confidence limits in the results (rather than the "wishy-washy" comments that permeate this report).

VII. Atmospheric Processes
1. pp.30-35: weak description of transport and mixing, and Appendix B is really quite poor.
2. p.42: bandwagon H2O2 oxidation? But let it pass. Appendix A is well done.
3. pp.44-53: in general, a weak description of the deposition processes, but acceptable in view of Appendix C.
4. p.50, 4L from bottom: No! "The motion of a molecule or particle ... by diffusion" does NOT depend on the gradient in concentration.

VIII. Theoretical Models
1. pp.55-68: This is really pathetic. Rather than all those words, how about showing some results?
2. p.70: "Dry deposition of NO2 differs somewhat from that of SO2 and is sometimes taken to be as little as on half that of SO2." I should hope so! How about two orders of magnitude smaller?
3. p.71, end: "Considering the crudeness ... it is suprising that the linear models perform better in estimating the long-term average wet deposition of sulfate than in estimating the ambient concentration."
What linear models? WHAT REFERENCE?!!

4. p.72, last paragraph, and continuing on to the end of this section, on p.75.
HOLD EVERYTHING. STOP. NOW WAIT A MINUTE. YOU GUYS HAVE MISSED THE POINT. THAT'S NOT THE LINEARITY VS. NONLINEARITY ISSUE!!!!!!!!!!!!!!!!
What are you guys, chemists by any chance? Familiar with reactions in test tubes, I presume. Oh yes, reactions can be linear or nonlinear in the concentrations. But suppose one of the reactants isn't present. Suppose you wanted to oxidize SO2 but there is no oxidant. Whether the reaction is linear or nonlinear (your meaning) in the oxidant is beside the point: the reaction ain't gonna occur if there ain't no oxidant! That's why the term "saturated" is used: the system can be said to be saturated with SO2. And in the atmospheric case, the system can then be nonlinear -- but it's not nonlinear in concentrations, it's then nonlinear in space. The system may be linear in the concentrations, but if there's nothing there to react, the system will respond nonlinearly: reduce emissions in the U.S. and the deposition in Greenland will be the first to start to decrease. Continue, and eventually there would be a response in Canada and then in the U.S. It's not a well-mixed test tube! (And of course I don't know if this will occur. But you don't know either -- so say so.)
5. pp. 75-82: And away you go with your focus on homogeneous, gas-phase chemistry. By any chance did the chairman write this section? There's a hint on p.78: "The best available experimental evidence today ... (Stockwell and Calvert 1983)!
And then did someone on the Committee do his best to rein him in? Page 82, complete with its uneven typing, leaves me with the distinct impression that somebody (Volker?), upon reading what was written, tried to holler "Chemists, whoa!":
"Note that for the case of an oxidant-limited reaction, a nonlinear response in sulfuric acid deposition WILL RESULT <emphasis added> from emission reduction....
"Limitations of oxidant for HSO3- or SO2 BECAUSE OF PHYSICAL PROCESSES <emphasis added> as well as the chemical influences described....
"Several types of nonlinear effects may be expected from factors NOT IMMEDIATELY RELATED TO OXIDANT LEVELS <emphasis added>....
"...we judge that nonlinear effects....probably small FOR THE GAS-PHASE CONVERSION STEPS <emphasis added>...
And then, after saying: "However, these conditions cannot be tested from the existing data base", why not make one additional, obvious statement? Something similar to: "Therefore, we really don't have the foggiest idea whether the system is linear or nonlinear (saturated or not)."
Instead, there's this (p.82, 3L from bottom): "Significant nonlinearity is anticipated for certain special conditions..."
LIKE DURING RAIN, PERHAPS?!

Findings and Conclusions.
6. p.83, end of second para: Here's another case of incredible logic. Follow this.
(i) "However, because of .... we do not regard currently available models as sufficiently developed to be used with confidence in predicting responses of the atmospheric system to alternative control strategies."
(ii) Those models have taken very competent people decades to develop.
(iii) But we have a few weeks here, so we'll develop a model of our own, and tell the nation how to control emissions. (YES, THEIRS IS A MODEL: NO DATA SUPPORT THEIR CLAIMS.)
7. p.84, top: What's this about the Rodhe et al. model being "widely used."? Who else is using it?
8. p.85, middle. A KEY COMMENT. "Measurements of oxidant concentrations in cloud water, although limited, suggest that concentrations may be sufficient in eastern North America for complete oxidation of SO2, except in winter."
JUST SLIPPED IT IN! NEVER MENTIONED BEFORE. NO REFERENCES. TOTALLY UNSUBSTANTIATED! BUNK!! WHO HAS MADE A SINGLE, RELIABLE, MEASUREMENT OF H2O2 IN CLOUD WATER? WHY IS AL LAZRUS NOW TRYING SO HARD TO PERFECT HIS METHOD? WHO SLIPPED THIS SENTENCE IN???
9. p.84, next sentence: "If this is the case, then strong deviations form linearity in the relationship between changes in annual average ambient SO2 concentrations and changes in the net production of sulfate in clouds would not be expected."
Oh? And why now "annual averages". And what about the fact that typically 50 percent of the deposition occurs during about 15 percent of the storms? And what about winter-time deposition? And what about the acidity from HNO3?
AND WHY? WHY ARE YOU GUYS DOING THIS? DO YOU THINK THAT YOU HELP THE PUBLIC BY DESTROYING WHAT LITTLE CONFIDENCE THEY MIGHT STILL HAVE IN THE INTEGRITY OF SCIENTISTS? WHY NOT STICK TO WHAT IS KNOWN? DON'T YOU THINK THAT YOUR COLLEAGUES ARE GONNA HOLLER "BUNK". THEY MUST, FOR AS MUCH AS THAT WILL HURT SCIENCE, NOT HOLLERING BUNK WILL HURT SCIENCE EVEN MORE.

IX. Emperical Observations (Gasp') and Source-Receptor Relationships

NOTE TO THE READER: My 40-hour weekend is now drawing to a close; I must move more rapidly, and then tomorrow, get back to working on what I'm paid to do.

1. There isn't much on pp. 89-99, anyway.
2. p. 99, bottom: "A correlation analysis of sulfate particle concentration in dry air between various SURE sites indicates that significant regions of correlation generally extend 200 to 300 km from major sources and rarely beyond 500 km (EPRI 1981). The exception to this observation occurs during ducting of pollutatnts from southern regions to the Northeast by southwesterly air."
That could be an interesting result, but it isn't: the correlation is undefined (What phase lags were examined? How was this related to meteorology?), and the exceptional case is almos. the only case of interest -- leading to acidic rain!
3. p.102, L6: Where did the Committee get the result: "In eastern North America, the warmer months tend to be the period of greatest precipitation ..." A grey-literature reference that happens to be sitting on my desk says: "The toal precipitation collected at the three MAPSS sites (during 1977 to 1979) had a winter/summer ratio of 1.06."
4. And more on p. 102, second to last sentence in this same paragraph: "These cases (European data) demonstrate that gas-phase photochemistry may not be the dominating factor contolling the seasonal trend of wet deposition of sulfate."
What a strange way to put it? There wouldn't happen to be an advocate of gas-phase photochemisty on the Committee, would there?
5. And after all that statistics, it's neatly summarized on p.113, end of section: "However, empirical orthogonal-function analysis (they love that word empirical) and other statistical techniques have not progressed to the stage at which they can be used to predict consequences of slective changes in emissions.
6. And so too with tracers (pp. 113-116). Therefore, left with no science, the Committee boldly extends itself into the realm of hand-waving arguments!

SO HERE IT COMES, PEOPLE: 116 PAGES INTO THE REPORT, ALL SCIENCE HAS NOW BEEN DISMISSED, AND IN THE NEXT 138 - 116 = 22 PAGES, YOU WILL SEE (IF YOU'RE CAREFUL) HOW SCIENTISTS, CHOSEN BY THE NATIONAL ACADEMY OF SCIENCE, MAKE 100 BILLION-DOLLAR DECISIONS!

7. p.116, 16L from bottom: "The central question is whether the data can be used to judge whether deposition is linearly or nonlinearly related to emissions.
NO FELLAS, GO BACK AND LOOK AT YOUR CHARTER; THAT WASN'T THE CENTRAL QUESTION. MOREOVER, IT WOULD BE GREAT IF YOU GUYS KNEW WHAT WAS MEANT BY LINEAR VS. NONLINEAR, IN THE CONTEXT OF SATURATION.
8. p.117, L12: what a dandy way to put it. I commented on this before. Watch the location of the word "only" in this sentence:

"The body of European data provides the only direct evidence for this phenomenon." <of nonlinearity> Never mind that the European data is the ONLY data worth a damn.
9. p. 117, bottom: and so what does the Committee plan to use? The Hubbard Brook data. BULK SAMPLES (That means that the sampler is open all the time, to provide a toilet for the birds, a swimming pool for bugs, a collector for leaves.....), COLLECTED ON A WEEKLY BASIS (why not: if Mother Nature didn't like weeks, then why did she invent weekends?), AT A SINGLE SITE ("No, the old power plant used to be over there, I think, and you know they closed that road over there in 19 ought 76, or so, and I remember the time when we didn't have these motorbikes,..."!)
10. p. 119, a little past the middle; hang on: "A linear-regression analysis of the Hubbard Brook data <I'm glad that these chemists can at least use their pocket calculators> against time indicates..."
and then (last paragraph)

"The Hubbard Brook record ... appears in toto <a nice touch> to demonstrate a reduction in sulfate concentration <okay so far> SIMILAR TO THE GENERAL REDUCTION IN SO2 EMISSIONS...."

Hello. Why did you slip that one in? I mean, what about a inverse relationship with the cost of gasoline? How about inversely with the distance of the Pioneer spacecraft from earth? WHAT THE DEVIL ARE YOU GUYS DOING? IS THAT ALL IT TAKES TO SATIFY A CHEMIST THAT THERE IS A RELATIONSHIP BETWEEN TWO VARIABLES? YOU'VE GOT TO BE KIDDING!
11. p.123, 3L from bottom (just for fun): "Obviates the usefulness of these data." Cute. Meaningless, but cute.
12. p.126, bottom. You know, a statement like the following could really upset a person. "The most reliable data available in a continuous record are those from Hubbard Brook. These data comprise the direct historical evidence for an emissions-deposition relationship <from Farmer Brown's tractor to ...> and show no indication of a significant nonlinearity." But why get upset. After all, these chemists are right: these data show no indication of a significant nonlinearity. Of course, they don't give any indication that the system is linear, either. But they do show that the bird droppings ...

WHAT, PRAY TELL, IS YOUR TEST FOR "SIGNIFICANT NONLINEARITY?" IN FACT, WHAT IS YOUR TEST FOR ANYTHING. WHAT DO YOU DO? PUT A FEW CURVES ON THE SAME PAGE AND SEE IF THEY'RE GOING THE SAME WAY?!

It appears so, and therefore (to devestate your only other basis for suggesting that the system is linear) I would have you make one more plot:

12. pp. 127-131. Here's what I want you to plot. Take the molar ratios for Europe (given in Table 4.7) and plot them against distance, say from your Region 1, and then add the data for N. America (your Fig. 4.17, p.130). Sure, it'll be crude:

Station	Distance (deg. lat.)	Ratio (normalized)
Europe:		
1	0	1
2	4	1.2 or .7
3	5	1.2 or 1.3
4	8	1.8 or 1.3
5	14	2.0 or 3.8
N. America:		
PA	0	1
Adirondacks	2	1.2
Quebec City (Your 1.6)	5	1.6

OKAY, NOW PLOT THEM. NOW, WHAT WERE YOU GUYS SAYING ABOUT CONSTANT MOLAR RATIOS??????????? This is so bad that even if I weren't out of time, I don't know how I could continue. Pure unsubstantiated bull! The opposite conclusion to theirs is staring them in the face!

Quickly, to the end:
13. p.133, middle: point measurements are useless; you already told us that you can't identify local contributions.
14. p. 133 bottom, 135 top: the area-wide average removes the important spatial variation.
15. p.135, L.6: "No retardation of SO2 oxidation ..." B.S. Plot the above table.
16. p.135, next paragraph: "Net oxidation of SO2 is as efficient.." B.S. Look again at Pack's paper, and then why the difference in summer vs. winter?
17. p.136, top: "No evidence of strong nonlinearity..." B.S. How about the spatial trend (which you chose to ignore) and the seasonal pattern?
18. p.136: I think my opinion about your inference need not be detailed.
19. p.136, middle: "If dry deposition is linearly proportional to emissions (as suggested in the section on principal-component analysis), then ..." Sorry, fellas, that must have been in an earlier draft; it's not there.
20. p.136, middle: "Without greater confidence in deterministic models..."
And would you mind, terribly, defining what you mean by "confidence".
21. p.138, middle: "We lack .... to predict, either qualitatively or quantitatively..." Are all those words really necessary?
22. p.138, back to "empirical observations" and "seriously

nonlinear".
FINDINGS AND CONCLUSIONS
1. p.139,L1: Great: "empirical data". Hey, I just thought of at least one highly appropriate use of the word empirical: this is an empirical report! ("relying or based on ... without reference to scientific principles"!)
2. And so on it goes, repeating the gargage.

X. RESEARCH NEEDS
Well done, but please: "Direct empirical observations in the field", and how about suggesting the scientific approach. You know: the old business about hypothesis, test, hypothesis, test,... Surely you jest. You must have heard about it. Yah, it's a great way to cut down on the number of wild speculations. No, like .. Well, once I read this empirical report and it said.....

P.S. Someone slipped it in again:
p.149, buried, you can find: "A limitation of oxidant in winter is believed to be a princial cause of such nonlinearity." Shh. Don't tell the chairman.

CLOSING COMMENTS

I was asked to review this report. No one asked for my opinion abaout the issue. But I'd like to add it.

I don't disagree with the general implication of this report. If depositions are to be reduced, then it seems more reasonable to decrease emissions than to increase them.

My disagreement is with the details in this report. A group of scientists, representing us all, were asked to examine the issue and report their examination. They went astray into the linearity vs. nonlinearity issue, and there is no scientific basis for the conclusions they present.

My point is this. We don't know if the system is linear or not, but if emissions are reduced, depositions will likely follow ... eventually. Otherwise, what should we do?

Reviewer I

By and large, the Committee has made a useful contribution by bringing before the public--in rather the most effective fashion to date--findings of many years of research on the transport, transformation, and deposition of airborne pollutants. Nevertheless, we do have some differences of opinion relative to the Committee's conclusions, differences that are primarily matters of scale and degree. But at this stage of developing knowledge of these complex atmospheric processes, and at this point in the developing consensus for some as yet poorly defined legislative action, these differences could have far reaching consequences.

The NRC/NAS report makes a number of conclusions and recommendations which we believe are either not convincingly supported by the evidence presented, or for which we prefer alternative views supported by the evidence of our own work. These include statements such as:

1) On an annual time scale and over a 1000-km spatial scale, the formation and deposition of sulfate do not appear to be significantly nonlinear.
2) Current operational models of transport and deposition are inadequate for policy analysis.
3) Relative contributions of specific source regions to specific receptor sites are unknown.

Since each of these points deals in some way with the question of the relative "validity" of current models, it is difficult to discuss them separately. The following attempt to do so proceeds in the order presented above.

The NRC/NAS report limits its finding of "no significant nonlinearity" to the larger scales given in 1), above. Our reservations in this regard are concerned with the improved definition of these scales, specifically the determination of the shortest transport times and smallest distances below which neglecting nonlinearity could have important consequences. Time scales over which nonlinearities may affect policy should be accomodated in the design of emission control strategies. Clearly these scales are related to ecological and material effects; effects on lakes and watersheds do seem to be largely long-term--perhaps seasonal, as in snowmelt shock. But if shorter term ecological "insults" are found to be important in current research (for example, effects on vegetation at critical stages of growth), then the magnitude of time scales for nonlinearity would need reexamination.

The 1000 km spatial scale is much larger than the distances between many combinations of source and sensitive-receptor areas. The possibility of nonlinearity over shorter distances would therefore have to be considered for the consequences of potential emission control policies to be assessed accurately. Then too, even if a uniform rollback policy were adopted, all emission sources could not be uniformly reduced because of economic or equipment constraints. Instead, rollback amounts would probably be defined over some political entity of medium scale, such as a state. Current emission levels per BTU vary widely now, and some sources are much more amenable to emission control actions than others. For instance, if a plant has already installed a scrubber, little additional emission reduction could be required of that plant if it is to continue to operate. Thus, rollback could not be expected to be uniform even when totaled by state. In short, the NRC/NAS report appears to address the issue of deposition reduction by control legislation too narrowly.

We agree with the report's conclusion that nonlinearities are more likely to occur in winter, when many transformation and removal rates are slower and transport generally faster. But even this is a qualified agreement pointing up the issue of scale: specifically, over just what distances would nonlinearity be expected to limit the efficacy of rollback legislation in these--or any--weather conditions? The consequences of the interpretation of nonlinearity are too far-reaching to be based on the limited data used.

The evidence cited by the NRC/NAS report in support of their argument for linearity is a) the ~ 15-year record of weekly bulk deposition collected at Hubbard Brook, New Hampshire, b) historical SO_2/NO_x emission estimates , and c) the observed spatial patterns of molar ratios of SO_2/NO_x emissions and sulfate/nitrate wet deposition. An apparent trend of decreasing sulfate deposition at Hubbard Brook is associated with a trend of decreasing SO_2 emission in the Northeastern U.S. But there are several difficulties with this analysis:

1. No uncertainty ranges are placed on any of the emission totals. Since the SURE, MAP3S, and NOI inventories for a single year (1978) show differences in total emissions by states of 30 to 40 percent, the historical emission data used may be equally--or even more--uncertain. Such uncertainties would swamp the apparent effects of any actual emission change.

2. Trends in Canadian emissions are not considered at all; yet the Canadian contribution to bulk deposition received in central New Hampshire, i.e., at Hubbard Brook, is probably about 25 to 40 percent of the total.

3. The apparent decrease in sulfate deposition at Hubbard Brook largely results from reference to relatively high values recorded in the initial years (approx. 1965-1969). Since that time, there appears to have been little or no further decrease in sulfate deposition, even though emission inventories over subsequent years did continue to decrease. Meteorological variability could have accounted for this deposition record, even if there had been no change in emissions; or a single, not-too-distant source operating only in the earlier years may have gone unnoticed.

4. The decrease in the molar ratio of the sulfate/nitrate bulk deposition at Hubbard Brook also seems to result primarily from a high ratio recorded in the late 1960s followed by irregularly lower values in subsequent years. The Hubbard Brook data suggests a step function rather than a trend. The possibility of analytical error--or a change in analytical technique--could be considered.

5. The NRC/NAS report does not resolve the paradox of the apparent evidence for linearity in the Hubbard Brook data and the evidence for nonlinearity in the European data. The comparison between statewide SO_2/NO_x molar emission ratios and sulfate/nitrate wet deposition ratios used to support the linear hypothesis, is not conclusive. For example, some preliminary results of modeling sensitivity tests now underway at this Laboratory suggest that the sulfate/nitrate wet deposition molar ratio is relatively insensitive to the SO_2/NO_x emission ratio; thus a decrease in SO_x emissions may not be proportionally reflected in a subsequent decrease in sulfate wet deposition.

6. The discussion concerning whether or not the Rodhe model is adequate without a chain-carrying radical to continue the reaction, glosses over the limitation imposed by the fact that both versions of the set of equations used are essentially clear-air chemistry models, and are therefore not directly relevant to wet deposition processes.

With regard to present models, we would suggest that the relative utility of many recent assessments indicates that a number of the numerical simulations now in use have been demonstrated to be adequate for policy analysis. Future improvements in modeling now proposed (and evidently advocated for future use in assessment by the NRC/NAS report) are more likely to improve diagnostic details and field experiment design than contribute directly to improvements in policy assessment exercises. Yet the report recommends against the use of current transport and deposition models for policy assessment, thus recommending , in effect, that no cost/benefit assessment be done at this critical time of legislative decision. These negative views on present modeling capability seem to be based upon the level of parameterization necessary for tractable models, and upon the Committee's perception of the results of the now outdated MOI model intercomparison.

Taking the second point first, there were many uncertainties in the 1978 SO_x emission inventory for North America used by the MOI comparison. Further, the precipitation chemistry samples used for model verification were monthly collections and were almost entirely derived from southeastern Canada. Finally, most of the models compared had only two months of 1976 meteorological data available to them. It follows that the MOI model intercomparison was hardly a rigorous test, especially in view of the improved data sets (not to mention the improved models) available today. The NRC/NAS report does concede that some present models "quantitatively point to the importance of certain geographical groupings of SO_2 sources and the potential influence of (those) sources on certain receptor areas". It can be added that some of today's parameterized models consistently reproduce, in realistic fashion, average (monthly and longer) downstream deposition patterns attributable to large groups of sources, in the form of so-called source-receptor matrices. A numerical model developed and used at this Laboratory, for example, achieves this degree of success with objective parameterizations of mixing, transformation, and removal processes which are based on actual measurements made in ad hoc field experiments. In other words, the model has not been arbitrarily tuned to attain this performance.

This brings us to the third point of difference. On the basis of our modeling experience, we believe that average monthly and seasonal contributions of specific source regions of significant size (say, half the area of a medium sized state) to receptor regions of similar size up to 1000-1500 km downstream, can be simulated with present models sufficiently well to make objective assessments. But until the emissions from particular source regions can be identified in deposition samples, or until large scale field experiments can be performed with realistically [illegible]/depositing tracers, no transport/deposition model of source/receptor relationships can be rigorously verified. Present models use real fields of meteorological data, after the fact. Indeed, it is recognized that whenever possible, assessments over a certain period (month, season) should be based on models run with several years of data in order to limit the effects of meteorological anomalies. We do not believe that there is any realistic need for a truly dynamic transport and deposition model that would operate in a meteorologically predictive mode.

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Telephone 312/972-5807

12 August 1983

Dr. Jack Calvert
National Center for Atmospheric
Research
P. O. Box 3000
Boulder, CO 80307

Dear Jack,

By now you have received the batch of reviews of the Acid Deposition report identified as from the National Laboratory Consortium. The reviews are from various individuals at the labs, and are not a unified product. In other words, no one at ANL is responsible for the review that you must have found personally insulting, and I regret that it was sent. Our review stressed our differing opinion as to the value of current models of transport and deposition, and to the definitiveness of the evidence cited for demonstration of linearity. Indeed, you have heard some of my opinions already.

I am enclosing some recent comparisons of ASTRAP simulations with observations at approximately 100 MAP3S, NADP, CANSAP, and APN sites. The correlations for nitrate deposition surprise even me. The dividing line between east and west is approximately the Mississippi River.

Here I have substracted from the observations an assumed natural background of 2.5 micromoles sulfate per liter and 1.0 micromoles nitrate per liter. The background assumption can change the bias but it has very little effect on the explanation of variance. I suspect the background concentrations are larger in the arid west, particularly if they are the result of dustfall contamination, or a seasalt sulfate.

Sincerely,

Jack

Jack D. Shannon
Atmospheric Physics Section
Environmental Research Division

JDS:mf

Enclosure

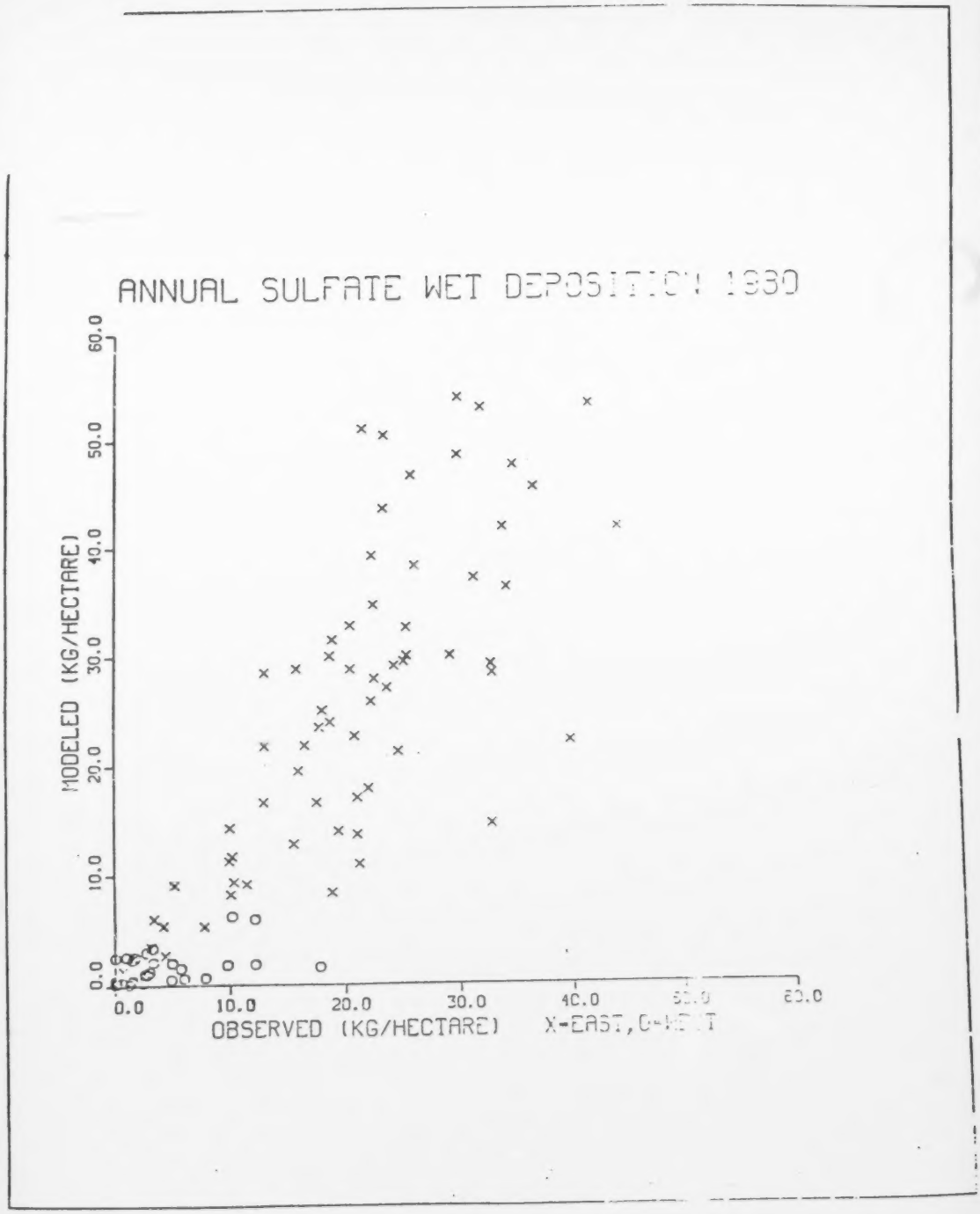
ASTRAP VERIFICATION STATISTICS

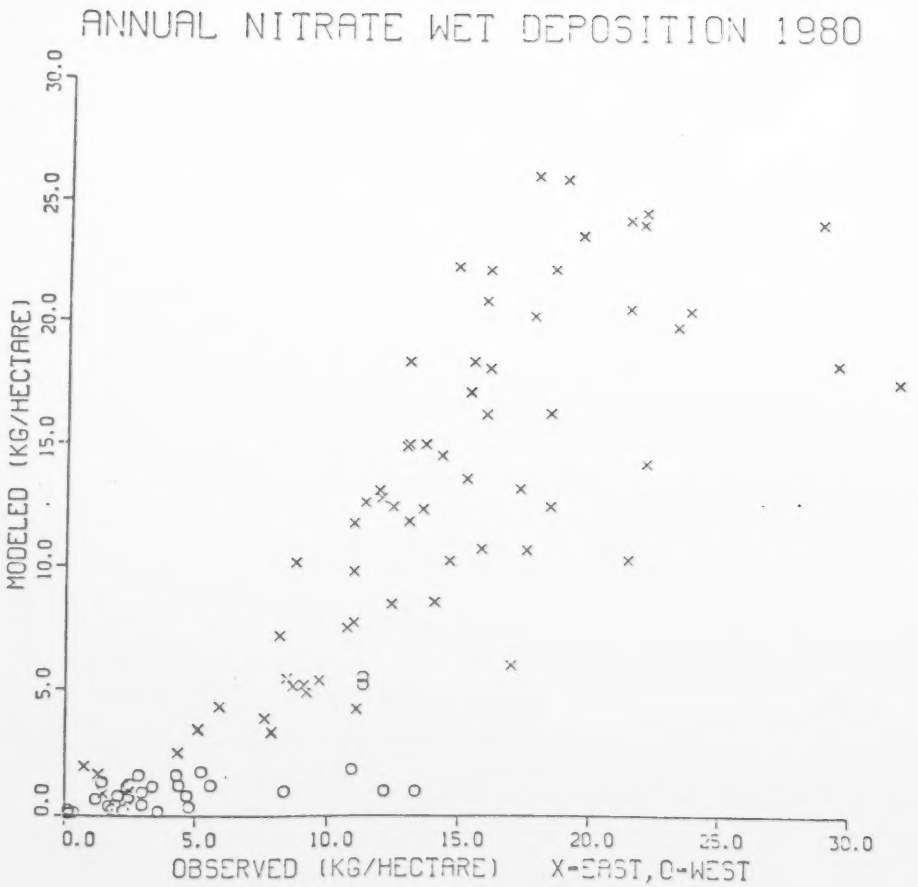
ANNUAL WET DEPOSITION 1980

	N	$\overline{OB}$*	$\overline{MODEL}$*	$\overline{RESIDUAL}$*	CORRELATION	VARIANCE EXPLAINED (%)
SULFATE						
ALL	95	18.1	20.6	-2.5	0.85	72
EAST	66	23.5	29.0	-5.5	0.75	56
WEST	29	5.9	1.7	4.2	0.35	12
NITRATE						
ALL	95	11.2	9.2	2.0	0.86	74
EAST	64	14.5	13.1	1.5	0.78	60
WEST	31	4.3	1.1	3.2	0.61	37

*UNITS KG SULFATE/HECTARE OR KG NITRATE/HECTARE

28-916 O - 84 - 51


ANNUAL SULFATE WET DEPOSITION 1980
MODELED (KG/HECTARE)
0.0
10.0
20.0
30.0
40.0
50.0
60.0
OBSERVED (KG/HECTARE)
X-EAST, O-WEST


ANNUAL NITRATE WET DEPOSITION 1980
MODELED (KG/HECTARE)
0.0
5.0
10.0
15.0
20.0
25.0
30.0
OBSERVED (KG/HECTARE) X-EAST, O-WEST

Revision of Appendix C: Comments of Jack G. Calvert and Alan Lazrus on the Informal "Review" of the NRC Committee Report on Acid Deposition Submitted by Bernard Manowitz Group

Recently we received a copy of a "review" of our NRC acid deposition report which was prepared under the guidance of Bernard Manowitz (Brookhaven National Laboratory) by selected representatives of three national laboratories supported by the Department of Energy: Brookhaven, Battelle Pacific Northwest, and Argonne. These "reviews" were sent to the National Academy of Sciences, to me, to various members of the Interagency Task Force on Acid Rain and others; my friends in industry and government tell me that the report was widely circulated to the leadership and research arms of the power industry and related industrial operations throughout the United States and within the extensive communications pipeline of the administrative and legislative staffs of the Federal and State governments. The original report has subsequently been sanitized and presented to Representatives Fuqua, Scheuer, and Lloyd with additional comments from a group at Oak Ridge National Laboratory; this has been or will be introduced into the Congressional Record. We did not have access to the revised version of the "review" of the Manowitz group until very recently, so we will necessarily respond to the original "review" which has received wide circulation. The scientific questions remain unchanged in the latest version. I have enclosed in this report a copy of the original Manowitz group "review" in the Appendix D.

The comments given here represent those of only two of our NRC acid deposition committee, myself (Jack G. Calvert) and Alan Lazrus, since time did not allow the participation of the entire committee. A Congressional hearing or the Congressional Record is not well suited to scientific debate. In the long run scientific issues cannot be influenced by legal or legislative actions

or by pressures from business, conservation groups or other special interest groups, including that headed by Manowitz. Indeed the scientific world does not accept blindly the stated opinions of any scientist or group of scientists, including the NRC committee on acid deposition, without extensive testing and checking of the ideas proposed. Fortunately for science the issues will be decided after extensive research and suitable discussions in the refereed scientific literature. However, I feel that the Manowitz group and its "review" cannot be disregarded by me since it has been presented to a wide and important audience as a scientific endeavor which disputes some key aspects of the NRC acid rain report. Although it is difficult to sort out the science from the rhetoric in some of these "reviews", we will focus on what we judge to be the important scientific questions which have been raised.

First, our use of the Hubbard Brook acid deposition data and the subsequent conclusions which we derived from them have been questioned. It is claimed by reviewer D that he has treated these and other data not available to the scientific community, in a more rigorous scientific fashion that did we. He concludes from this treatment that there is no 1:1 proportionality between sulfur oxide emissions and sulfate deposition at Hubbard Brook. Let me say at the outset that reviewer D is working with a set of data which have not been available to our committee. We had access only to the monthly averages which were reported in the preliminary writings of the Interagency Task Force on Acid Rain. Furthermore, we received only a preliminary version of the recent study edited by Munn of the University of Toronto (1982); this was designated by reviewer D as part of the extensive discussion and analysis of Hubbard Brook data available today, although it has not been published or released to our knowledge and appears to be in very rough form. We were requested not to quote or use this report in our considerations because of its preliminary nature.

Our response to reviewer D's comments are not altered by these complicating factors.

The arguments of reviewer D are seriously flawed scientifically for several reasons. He does not provide any evidence that justifies the variables employed in his multiple linear regression analysis; in fact, one of these was clearly inappropriate (his "wave" function). In reviewer D's treatment the actual observed sulfate values have been radically altered by "theoretical" adjustments of the original data. These adjustments are simply the results of multiple linear regression equations using three variables: the period of the year, surface temperature at a site miles from Hubbard Brook, and the amount of precipitation at Hubbard Brook. The corresponding regression coefficients, for which statistical significance is not demonstrated, are ambiguous.

a) Although the time of the year affects the extent of sunlight and oxidant generation, it has also been shown in work by Tanner and Leaderer (1982) to influence the contribution of long-range transport to sulfate in aerosols.

b) Though there is only one correction term for precipitation, it has been clearly demonstrated at Hubbard Brook that different types of storms manifest different relationships between the amount of precipitation and the concentration of sulfate. See the NRC report on acid deposition, p. 110-111. Increased precipitation during convective storms tends not to significantly decrease sulfate concentration, while in the case of warm frontal precipitation, sulfate concentration is inversely related to the volume of precipitation because of dilution (Munn, et al., 1982). On a climatic basis, an increase in frequency of storms may not have a significant impact on concentrations, whereas larger storms, depositing more water, may be inversely related to sulfate concentrations. A single regression coefficient cannot meaningfully account for these complex effects.

c) The temperatures employed by reviewer D are monthly averages collected at a location 75 miles from Hubbard Brook. Data from weekly precipitation samples were regressed against monthly averaged temperatures, even though there is no model based on physical and chemical principles which is capable of realistically relating surface temperatures to the incorporation of sulfate in rain.

The square of the correlation coefficient is a measure of how much variance in sulfate concentrations is attributable to the variance of sulfur dioxide emissions. Reviewer D's treatment of the data appears to be an attempted mathematical reduction of the variance of the data, with the likely intent of removing portions of the variance not dependent on sulfur oxide emissions. However the parameters selected to accomplish this goal are themselves not physically unrelated to sulfur oxide emissions or to the transport of sulfur oxide emissions to remote sites. The data adjustments applied by reviewer D are on the one hand likely to remove some variance related to sulfur dioxide since the parameters used are related to sulfur dioxide emissions. On the other hand if these are inappropriate variables for the adjustment, then they will lead to nothing but an increase in the error component in the adjusted sulfate record. One must question the effectiveness of the adjustment when the mean for the adjusted record is 42% greater than that actually observed at Hubbard Brook. It is very important to note that reviewer D's weighted estimates of annual averages of sulfur dioxide emissions do correlate at the 95% confidence level with the annual average sulfate concentrations reported by Likens. After "adjustment" of the actual data by reviewer D, there is no correlation. The correlation coefficient is no longer statistically different from zero or negative values. The data "adjustment" is not justified.

In our view there is a second major error in reviewer D's statistical treatment. In order to "correct" the observed data he models the long-range

transport of sulfur dioxide in a most unrealistic way. Based upon the failure to detect a strong correlation between local wind direction and sulfate observed in precipitation at Hubbard Brook, reviewer D surmised that sources from all directions can effect Hubbard Brook depositions equally and that their impacts are functions of the source strength and source distance alone. This is not an acceptable hypothesis. Furthermore reviewer D has chosen to use as representative of Hubbard Brook the local wind directions from a distant station on the coast of Maine, about 100 miles from Hubbard Brook, New Hampshire. A great many studies have shown the error in the use of even the actual local wind directions as an indicator of source region for long-range transport. In fact, the calculation of back trajectories of air parcels which bring precipitation to Hubbard Brook, suggest that less than 10% of the air masses come from the Midwest or EPA Region V (Munn, et al., 1983). Since the emission rates for sulfur dioxide in Region V are very much greater than the emission rates for regions I or II (New England), but the distance from Hubbard Brook to the centroid of emissions in Region V is only several-times greater than that of regions I or II, the contribution of the Midwest in reviewer D's model is comparable to that of the Northeast. This is totally inconsistent with the actual air parcel trajectory analyses. Since SO_2 emissions in Region V vary differently with year than in Regions I and II, the correlation analysis will be distorted. Thus the lack of statistically significant correlation between reviewer D's calculated sulfur dioxide parameters and any other variable is quite meaningless in view of Munn's trajectory study. Furthermore both the NRC acid deposition committee report and the full report of the Interagency Task Force on Acid Rain state that no present model describes in a quantitative fashion the relative impact of sulfur dioxide sources as a function of their strength and distance. Given the lack of such reliable models, it is not scientifically sound to attempt the treatment of SO_2 emissions and sulfate

concentration at remote sites given by reviewer D.

The conclusion of the NRC committee, namely that a large decrease in sulfate deposition at Hubbard Brook corresponded in time with a large decrease in sulfur dioxide emissions in the northeast, is as definitive a statement as was permitted by the data. Certainly the attempt to be more quantitative made by reviewer D is seriously flawed scientifically, and it must be rejected.

His treatment of sulfate to nitrate ratios which appears in the revised version suffers from the same problems, and the results are of uncertain quality. Reviewer D is critical because of the inexact correspondence between the molar ratios of SO_2/NO_x in emissions and $SO_4^=/NO_3^-$ in wet deposition. However the report recognizes the discrepancies and deals with them in the following excerpt from this report:

> It is surprising to note in addition that the annually averaged molar ratios in precipitation at the HAP3S, NADP, and CANSAP sites in eastern North America resemble the molar ratios of SO_2 to NO_x emissions averaged on an annual basis by state or province (Figure 4.18). The emission ratios are somewhat higher than the deposition ratios in the Ohio and Tennessee River Valleys. Similarly, the emission ratios are somewhat lower than the low deposition ratios immediately north of Lake Ontario. The smaller spatial variability of the deposition ratios compared with the emission ratios near large point sources is probably related to a rapid rate of atmospheric mixing relative to the rates of wet-deposition processes. Alternatively, a preferential dry deposition of SO_2 relative to that of NO_2 may occur in the near-source region, or the differences may indeed represent a suppression of SO_2 oxidation within a short distance from the emission sources. However, the general similarity of wet-deposition and emission ratios on an annual average basis suggests that the incorporation of sulfuric acid in rain is not retarded with respect to the conversion and incorporation of nitric acid in rain.
>
> A small body of data suggests that the historical trends in the molar deposition ratios of sulfate to nitrate correspond to the trend in molar SO_2/NO_x emission ratios. The historical trends of the molar deposition ratio $SO_4^=/NO_3^-$ at Hubbard Brook and the molar ratios of SO_2 to NO_x in emissions in the eastern United States by EPA Region are plotted in Figure 4.19. Also shown in the figure is the result of a linear-regression analysis of the Hubbard Brook deposition ratio against time. Between 1965-1966 and 1979-1980, the linear regression gives a slope of -0.039 (standard error 0.006) and an intercept of 1.444 (standard error 0.054). The significance level of the slope is 0.0001.

Another perspective is provided by comparing the annually averaged regional wet-deposition rates with regional emissions. Integrating wet deposition over the northeast quadrant of the United States [using the isopleths of Pack (1980) based on MAP3S and SURE data] Golomb (1983) found sulfate deposition to be 4.3×10^{10} (±20 percent) moles/year and total nitrate deposition to be 3.9×10^{10} (±25 percent) moles/year. Golomb calculated regional emissions from U.S./Canada (1981) to be 2.2×10^{11} moles/year of SO_2 and 1.8×10^{11} moles/year of NO_x. The emissions were similarly distributed geographically. Golomb also indicates that 19 ± 6 percent of the sulfur and 20 ± 7 percent of the nitrogen emitted in this region are wet deposited there. It is interesting to note that Golomb found the molar ratio of sulfate to nitrate wet deposited annually in this region to be 1.1, compared with an annually averaged molar ratio of emissions for SO_2 and NO_x of 1.2. The similarity of the ratios suggests that, whatever specific processes are taking place, there is a tendency on an annual basis for sulfate and nitrate to be deposited in the northeast quadrant of the United States with no significant loss of one component relative to the other.

When Reviewer D realizes that atmospheric mixing may be important, he resorts to simplistic hypothetical considerations of transport which are relatively meaningless.

Reviewer D is concerned with the large discrepancy between emissions and deposition ratios in the northern Canadian provinces. However, these large emission ratios (e.g., 30.96 for northern Quebec) are associated with only 560×10^{-3} tons of SO_2 per year in a region of about 480,000 square miles. In terms of average annual emissions of SO_2 per unit area, the northern Quebec region resembles Nebraska. The report restricted its comments regarding ratios to the region bounded on the north by the acidity isopleth of pH = 4.6. At lower acidities, it is not possible to discriminate between the effects of regional anthropogenic sources, imported sources, and natural sources on the $SO_4^{=}/NO_3^{-}$ ratio. The report maintains only that within the region of precipi-

tation acidity higher than 4.6, i.e., clearly anthropogenic, there is no evidence of large scale nonlinearities.

Reviewer D maintains that an insignificant correlation exists between weekly $SO_4^{=}/NO_3^{-}$ ratios at Hubbard Brook and SO_2/NO_x emissions in Federal Regions I and II. It is not statistically feasible to compare weekly deposition ratios with annual emission ratios. If reviewer D adjusted the weekly values to obtain annual averages as he did in the case of sulfate concentrations, the results are of questionable relability.

Review E has written his comments with such an extraordinary degree of rancorous disdain that we are tempted to neglect his comments completely. The reviewer must have been driven by desperation to resort to such levels of distorted analysis. His approach is typified by the following salient argument copied from his review:

12. pp. 127-131. Here's what I want you to plot. Take the molar ratios for Europe (given in Table 4.7) and plot them against distance, say from your Region 1, and then add the data for N. America (your Fig. 4.17, p.130). Sure, it'll be crude:

Station	Distance (deg. lat.)	Ratio (normalized)
Europe:		
1	0	1
2	4	1.2 or .7
3	5	1.2 or 1.3
4	8	1.8 or 1.3
5	14	2.0 or 3.8
N. America:		
PH	0	1
Adirondacks	2	1.2
Quebec City (Your 1.6)	5	1.6

OKAY. NOW PLOT THEM. NOW, WHAT WERE YOU GUYS SAYING ABOUT CONSTANT MOLAR RATIOS????????????
This is so bad that even if I weren't out of time, I don't know how I could continue. Pure unsubstantiated bull! The opposite conclusion to theirs is staring them in the face!

Region I was selected by Rodhe et al. to represent the industrial source region, and in fact comprises the Ruhr Valley and the intensively industrialized area to the West of it. The other regions were chosen to represent downwind areas in which the average chemistry of the precipitation would reflect changes in the chemistry of the pollution plume as a function of distance from the source region. It is assumed that pollution sources enroute are not large enough to introduce large changes in the plume chemistry. The NRC acid deposition report examines the molar ratio of sulfate to nitrate in each of the regions to discern whether rapid consumption of oxidant by nitrogen oxides in the source region retards the oxidation of SO_2 until the plume travels to greater distances. The observed ratio increases with distance consistently with this hypothesis, but as the report states, there are other explanations. The report states however that the lack of a gradient in ratios is not consistent with the hypothesis, and in North America no gradient is apparent.

Reviewer E chooses to normalize the ratio of sulfate to nitrate to unity at the source of the region with the simple proportion: X/region ratio = 1.0/source ratio. However, it is clear from Figure 4.17 that the ratio is not increasing between Pennsylvania and the Adirondaks, and his normalized ratio should be 1.0 for the Adirondaks, not 1.2 as he indicates. The nearest station to Quebec City indicates an observed average of 1.3, or a normalized value of 1.2, not 1.6 as Reviewer E indicates.

A more serious distortion, however, arises from reviewer E's use of central Pennsylvania as the principal origin of a regional plume causing acid rain over North America. A more realistic comparison with the source region selected by Rodhe et al. would be the Ohio River Valley. In the attached

copy of the NRC acid deposition report Figure 4.17, arcs are drawn from Cincinnati (a central location in the Ohio River Valley) corresponding to the linear distances between the center of each downwind region and the source region in the European map (Figure 4.16). Contrary to the table of reviewer E, the figure indicates that even at 8 degrees distance from the source region there is no indication of an increasing gradient of the ratio.

The NRC acid deposition report is limited to a discussion of $SO_4^{=}/NO_3^{-}$ ratios to regions where the pH is 4.6 or lower, since at higher pH values the ratio is probably influenced at least as much by local SO_2 sources, natural sources, or globally distributed, anthropogenic sources as by long range transport from the Ohio River Valley. At pH 5.0 the acidity is within the range of global background values (NRC acid deposition report, p. 13). The northernmost region in the European map was characterized by a $SO_4^{=}/NO_3^{-}$ molar ratio of 5.7, and was also typified by a pH of 4.6 in 1974 (Likens et al., 1979). However the analogous outermost arc on the attached version of Figure 4.17 lies at 1,150 miles from the source region. It falls in the region of pH 5.0 or higher in North America, and the ratio can no longer be realistically considered as reflecting principally chemical transformations in the original plume.

Referee E has implied that the gas phase chemistry is unimportant in acid deposition in northeastern North America. Sulfuric acid generation occurs in clouds and in gas phase reactions. Indeed evidence suggests that oxidation of sulfur dioxide in clouds in northeastern North America may be more important than that occurring in the gas phase. However the effective oxidants, ozone, hydrogen peroxide, and possibly other peroxy compounds, which are believed to be the necessary reactants for sulfur dioxide (bisulfite ion) in clouds, all are formed in gas phase chemistry. Thus in terms of current theory for clouds which have low concentrations of metal ion catalysts (Fe^{3+}, Mn^{2+}, etc.), if we don't

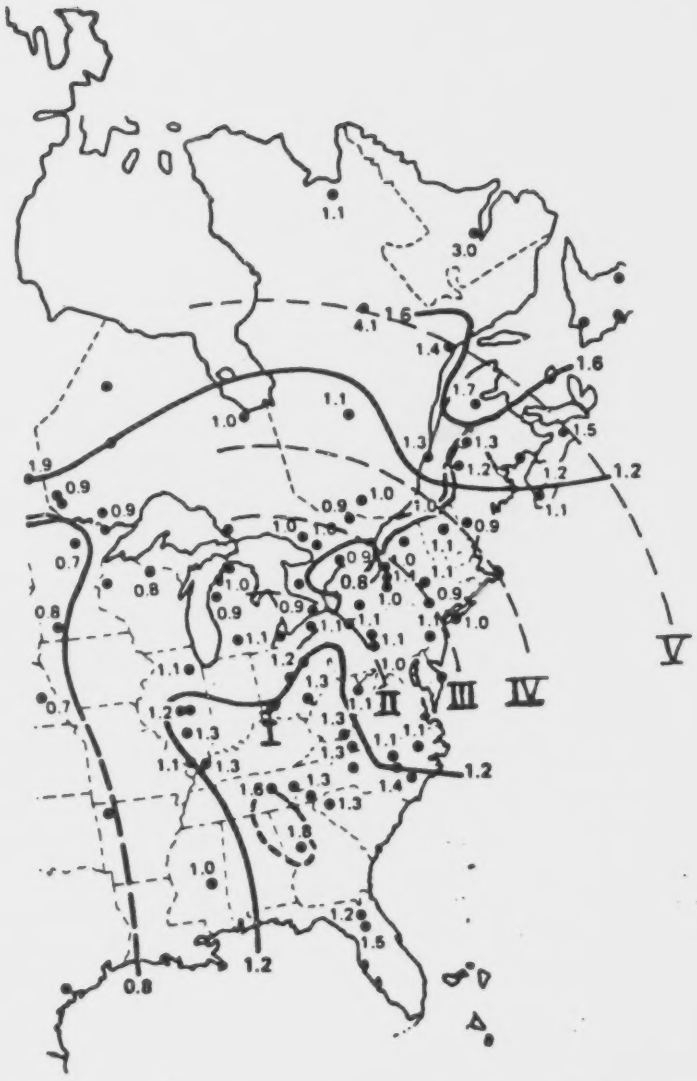


FIGURE 4.17 Average molar ratio of sulfate to nitrate in precipitation in eastern North America in 1980. SOURCE: B. Heikes, National Center for Atmospheric Research, personal communication (1983). Based on data from U.S./Canada Work Group #2 (1982). (Arcs drawn in the figure have been added to show the distance relationships which correspond to those used in the European map; Fig. 4-16.

FIGURE 4.16 Locations of monitoring stations used in the data analysis of Table 4.7.
SOURCE: Rodhe et al. (1981).

include gas phase chemistry we cannot account for the rapid rates of acid generation which have been observed to occur in clouds. Gas phase and solution phase reactions are highly coupled, and no scientist today would attempt to rationalize the chemistry involved in acid deposition without including both gas phase and solution phase processes.

One other important conclusion of our NRC acid deposition report has been questioned. We conclude that the reliability of long-range transport and transformation models is presently low in relating specific sources with receptor sites in sensitive areas. This conclusion does not seem to differ from that expressed in the report of the Interagency Task Force on Acid Rain (with which the Manowitz group has some undefined relationship), and the brief report of the Acid Rain Peer Review Panel. It also appears to be in accord with the comments of reviewer C (Bruce B. Hicks) of the newly added Oak Ridge "review" group. Work of Dr. Jack Shannon of Argonne National Laboratories has shown in very recent unpublished simulations using the ASTRAP model (private communication to Jack G. Calvert) that very good correlations are found for the calculated annual wet sulfate and nitrate deposition in eastern North America and the depositions observed in the 100 MAP3S, NADP, CANSAP, and APN sites for the year 1980. See the communication from Dr. Shannon in the attached letters in Appendix D. Such results look encouraging as related to long-time averages over a large area. It is interesting that a proportional or linear chemical model (with diurnally and seasonally varied rates of SO_2 oxidation), rejected by the Manowitz group, was employed in these seemingly successful long-term average simulations. A more important question at hand is how well present models can do in establishing the percentage of the deposition in a given sensitive area which can be ascribed to a given emission source and

the extent of cutback in acid deposition in sensitive areas which we can expect from control of certain large sources which are well-removed from the site. It is the opinion of the NRC committee on acid deposition, the Interagency Task Force on Acid Rain, as well as the President's Panel on acid rain that this cannot be done now with confidence. Models are used by all of us in the sciences today, and certainly definitive uses have been and will continue to be made of them. Our caution relates to their inappropriate use. If such modelling is used in control policy formulation at the present time, and, of course it is being used now, then the modeller should provide the user with realistic estimates of the magnitude of the uncertainty associated with the results so that these can be incorporated into the control strategy planning. Such a practice is not suggested to frustrate the control plan, but it is necessary to provide the sense of realism that is demanded in view of the very high costs involved in the contol of sulfur oxide emissions.

One final point should be made. The NRC report states that the Rodhe, Crutzen, Vanderpol model (1981) which was used to explain the possible nonlinearity between $SO_4^=$ deposition and SO_2 emissions should be modified in the light of recent specific laboratory findings (Stockwell and Calvert, 1983). Reviewer A doubts that this modification would significantly influence the result. It should be noted that the suggestion of the NRC report, supported by simulations of Professor Perry Samson of the University of Michigan which are given in the report, was tested independently in a comprehensive model of the gas phase chemistry of SO_2, NO_x and hydrocarbons by Hov (1983). It was found that complete regeneration of radicals in the SO_2 oxidation reactions as suggested by the laboratory study leads to a nearly proportionate (linear) response of reduced sulfuric acid formation to reduction in SO_2 emissions in long-range transport; 50% reduction in SO_2 gave a 48% reduction in sulfate generated.

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Certainly the conclusions of the NRC committee on acid deposition are necessarily very limited in nature, and indeed they must and will be checked by extensive further experimentation. Our conclusions represent the best judgment of a group of highly qualified and diversified scientists, and we certainly would not alter or compromise these conclusions on the basis of the unusual "review" of the Manowitz group.

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Attached in the next seven pages of this report is a discussion presented by Professor James Williams of the Department of Statistics, Colorado State University related to the statistical problems in treating the Hubbard Brook data.

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Comments on the "Review" of Manowitz referee D by Professor James S. Williams,
Department of Statistics, Colorado State University, Fort Collins

A. Linear and Nonlinear Systems; Linear and Nonlinear Regression

Members of the National Academy of Sciences study panel have focused on the question of whether or not emission-deposition processes for SO_2-SO_4 and NO_x-NO_3 are linear or nonlinear. Reviewer D contests the published conclusion by panel members that linearity is a workable approximation for those systems in North America that are like the one sample in the Hubbard Brook Experimental Forest in New Hampshire. The definition of linearity set out in the panel report (p. 73) appears to be the same one used by the reviewer in much of his analysis, discussion, and conclusions. However the definition is too restrictive, as noted (at least by implication) in both the report and review.

A linear system in real variables is one in which input variable $\underline{x}$ is transformed into $t(\underline{x})$ where the function $t(\cdot)$ is characterized by:

$$t(\underline{x} + \underline{y}) = t(\underline{x}) + t(\underline{y}) \text{ for all } \underline{x}, \underline{y} \in R^p$$

In such a system if input is changed from $\underline{x}$ to $m\underline{x}$, then output is changed from $t(\underline{x})$ to $mt(\underline{x})$. In particular if $\underline{x}$ is a numerical measure of man-made emissions and $t(\underline{x})$ is a measure of depositions, then reducing emissions to a fraction m of $\underline{x}$ would reduce depositions to the same fraction m of $t(\underline{x})$. If man made emissions were eliminated ($m = 0$), then depositions would be eliminated. This last result is unrealistic because undoubtedly there are depositions which are not the result of man-made emissions. This objection can be removed by considering other types of systems. Systems of the type just described are called homogeneous linear; a more appropriate type for consideration is nonhomogeneous linear systems for which:

$$t(\underline{x}) = t(\underline{0}) + u(\underline{x})$$

$$u(\underline{x} + \underline{y}) = u(\underline{x}) + u(\underline{y}) \text{ for all } \underline{x}, \underline{y} \in R^p$$

In such a system input results in a homogeneous linear change added to output associated with zero input. If input is changed from $\underline{x}$ to $m\underline{x}$, then the output is changed from $t(\underline{x})$ to $t(\underline{x}) + mu(\underline{x})$. In such a model for an emission-deposition process, the elimination of man-made emissions ($m = 0$) would not eliminate depositions, but would reduce them to a natural level ($t(\underline{0})$). Also the rate of change in output with respect to any particular input variable can be a constant other than 1, the only value it can be if the system is homogeneous linear.

A system which is not linear is nonlinear. It is characterized by rates of change which are not constants but instead depend on $\underline{x}$. For such a system, the change in output in response to a change in input can be calculated but it is more complicated in form than the change in output for a linear system.

The terms linear and nonlinear are also used to qualify different types of regression analyses. However, they are not used in this context in the same way that they are used to characterize a transformation of input into output. For example, the nonlinear transformation of a single input variable $\underline{x}$ into output $t(\underline{x})$ which is given by:

$$t(\underline{x}) = \beta_0 + \beta_1\underline{x} + \beta_2(1/\underline{x})$$

might be used as the basis of the model of linear multiple regression analysis. With respect to the regression analysis, linear applies to the manner in which the coefficients β_0, β_1, and β_2 enter the equation for $t(\underline{x})$ rather than to the manner in which $\underline{x}$ enters the equation. In general, the methods of linear regression apply to all linear transformations of input $\underline{x}$ into output $t(\underline{x})$ and either exactly or approximately to a very large class of nonlinear transformations. There are also methods of nonlinear regression analysis which can be applied to nonlinear transformations to replace approximate

procedures by exact ones and to enlarge the class of transformations of input to output that can be studied with a regression analysis.

The conclusion to be drawn from this discussion is that the conclusion presented by the panel members and contested by the reviewer is not as central as the former indicate and is not as restrictive to consideration of emissions control policy as claimed by the latter.

B. Statistical Analysis of Relationship of Deposition Data from Hubbard Brook Experimental Forest to Regional Emissions Data

Efforts to derive equations for the transformation of emission variables into deposition variables have not been successful enough to predict the latter in any given region from measurements of the former in nearby and more distant regions. One can predict with confidence that such equations will never be found because the system includes a number of random components which cannot be included in such equations. The successes of studies of this type will increase our knowledge of possible workings of tranport, chemical conversion, and deposition processes in the system.

Satisfactory predictions can be derived from the results of regression analysis if the more important variables affecting deposition can be identified and measured and if sufficient data can be collected to insure accurate results of a regression analysis. At the present time the complete list of important variables for even one deposition site, such as the Hubbard Brook Forest, is unknown, and only a modest amount of data are available for analysis. These two reservations are clearly stated in the panel report and are given as reasons for their basing conclusions on an informal analysis for qualitative descriptions of relationships. No attempts were made at formal analyses for quantitative descriptions and predictions.

The main difficulty one would encounter in a regression analysis of the relationship of the Hubbard Brook Forest data to the emissions data for the Mississippi valley and regions to the east can be seen in Figures 4.14 and 4.15 of the panel report. The modest trends present in the series of yearly deposition figures for $SO_4^=$, NO_3^-, H^+, and pH (Figure 4.14) are hypothesized to be responses to trends in the series of yearly emissions for five regions either nearby the Hubbard Brook Forest (Regions I and II) or more distant (Regions III, IV, and V). These emission trends contain strikingly different changes in direction for SO_2 and leveling out for NO_x. In each region there is a year of maximum SO_2 followed by a decline in levels. This occurs in 1965 for Regions II and V, in 1970 for Regions I and III, and in 1975 for Region V (Figure 4.15). The apparent decline in $SO_4^=$ deposit begins in 1968 (Figure 4.14). If it is a positive response to changes in emissions, then it must be a delayed effect of the turn downward in SO_2 emissions in Region II and/or Region V. If it is a negative response to changes in emissions, then it must be an indication of increasing dominance of Region IV on the level of SO_2 contamination reaching the Hubbard Brook Forest. In each region, the level of NO_x emissions either levels off or even declines slightly after a period of increase. This occurs in 1965 for Region III, in 1970 for Regions I, II, and V, and in 1975 for Region IV (Figure 4.15). The increase in NO_3^- deposition lasts until 1970 (Figure (4.14) and, therefore, is apparently a positive response to emission changes in Regions I, II, and V or a negative response to increasing dominance of Region IV on levels of NO_x contamination reaching the Hubbard Brook Forest. Also the possibility that effects on one source of acid deposition might be indirect, that is the consequence of changes in the other source, cannot be overlooked.

Summary indices of emissions from all sources, such as the totals for SO_2 and NO_x graphed in Figure 4.15 or the weighted totals for SO_2 and NO_x presented by Reviewer D, are inappropiate input variables unless they are constructed to emphasize by correct weighting, the real effects of the regions on the levels of contaminants in the air mass of the region where deposition is measured. Reviewer D has implied that weighting by either inverse distance or inverse squared distance is correct. However he presents no analysis to support this conclusion and his citation of the study published by Munn et al. is inadequate because the analyses prepared by that group are so few in number and unfocused.

An important point to realize is that the regression analysis cannot be used to identify sources of emissions which truly affect deposition variables in a region. However the analysis can be used to eliminate some of the regions which have negligible effects if the data record includes patterns in changes in emission trends which can be used to distinguish each region from all others. It is fairly certain that a long record would be needed to achieve this. It cannot be done with the available data because trends in Regions I, II, and III are similar for SO_2 and trends in Regions I, II, III, and V are similar for NO_x emissions (Figure 4.14).

Reviewer D has based his conclusion that yearly SO_4 depositions are not linearly related to SO_2 emissions on regression analyses in which an input index is used. An example of one of these indices is graphed in his Figure 2 along with a graph of adjusted SO_4 depositions in the Hubbard Brook Forest. The reason that there is no significant linear relationship is obviously due to the marked counter trends in the two series up to 1970. Were Region II to receive greater weighting in the index, much of the upward trend in the SO_2 trace would be eliminated, and a stronger positive linear relationship would

be obtained. Were Region IV to recieve greater weighting in the index, much of the downward trend in the SO_2 trace would be eliminated, and a strong negative linear relationship would be obtained. It is clear that the decision of how to weight regional emissions in the index determines the sign and the statistical significance of linear dependence of SO_4 depositions on SO_2 emissions. Were the correct weightings known, the regression analysis could be properly applied to determine if there is evidence in the data for linear dependence. Panel members in their report have argued that the correct weightings are not known and that as a consequence a formal analysis is not justified (p. 140-141).

C. Statistical Adjustment of Deposition Data to Eliminate Effects of Meteorological Variables

Reviewer D has used linear regression procedures to adjust deposition measurements taken at Hubbard Brook Forest for precipitation volume, surface temperature (at Concord, New Hampshire) and all other factors associated with cosine variation in seasonal trends. It is not clear whether the analysis for coefficients used in the adjustment was calculated from the data broken into yearly subsets (a within-year analysis) or from the unpartitioned records because the reviewer notes that coefficients are the averages of several coefficients (page 8). Presumably this comment refers to something else because only the latter method of calculation is appropriate.

The choice of cosine variable to represent cyclic trends is ineffective because year-to-year variations in seasonal effects which are necessary for a proper adjustment are not included in the cosine variable. The only consequence of inclusion of the cosine term could have would be to increase the residual variance of the adjusted records and thereby reduce the statistical significance of tests based on the adjusted data.

Both panel members and the reviewer note that summer deposition of $SO_4^=$ dominates the yearly record. Because of this, further analyses of $SO_4^=$ data should be done separately for the yearly records of depositions during warm and during cool seasons. Changes in the yearly deposition trend should show up more clearly in the warm season record than in the yearly record.

LONG RANGE TRANSPORT OF ACID RAIN PRECURSORS

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ABSTRACT

A model of the long range transport of primary and secondary pollutants derived by Fay and Rosenzweig (1) is applied to the problem of the transport of acid rain precursors. The model describes the long term average (annual or seasonal) airborn pollutant concentration due to a single source. Because the chemical transformation and physical deposition processes are assumed to be linear in the concentrations, the contributions of many sources may be determined by superposition. Simplified forms of the source-receptor relation are derived for ranges of the model parameters which are appropriate to sulfur oxide species. Quantitative results of applying the model to airborn sulfates in the eastern U.S. are compared with more complex models.

This work was partially supported by a grant from the M.I.T. Center for Energy Policy research. The author wishes to acknowledge the interest and helpful discussions of D. Golomb of the M.I.T. Energy Laboratory.

NOMENCLATURE

D	Horizontal turbulent diffusivity
f_{si}	Fraction of secondary pollutant concentration due to source i
h	Height of atmospheric mixing layer
k_{si}	Ratio of secondary pollutant concentration to primary source strength, Eq. (14)
K_0, K_1	Modified Bessel functions of order 0 and 1
ℓ	Length scale defined by Eq. (4)
Q	Primary pollutant source strength
Q_i	Primary pollutant source strength of source i
r	Radial distance of source from receptor
s	Length scale defined by Eq. (10)
w	Mean wind speed
x	Streamwise distance of receptor from source
y	Normal distance of receptor from source
β	Mass of secondary pollutant formed per unit mass of primary pollutant
λ	Length scale defined by Eq. (3)
η	Length scale defined by Eq. (8)
τ	Inverse of rate constant for loss of primary pollutant
τ_c	Inverse of rate constant for conversion of primary to secondary pollutant
τ_s	Inverse of rate constant for loss of secondary pollutant
X_p	Concentration of primary pollutant
X_s	Concentration of secondary pollutant
X_{so}	Concentration of secondary pollutant at $r = 0$

INTRODUCTION

Acid rain is the popular name for wet deposition to the earth's surface of acidic solutions in the form of rain, snow, sleet, fog, and mist. Acidic aerosols also are deposited slowly by gravity; this is termed dry deposition. The acidic content of these deposits is principally due to sulfates and nitrates, but other species of lesser importance may also be present. In the heavily industrialized regions of eastern North America and Western Europe where precipitation is most acidic, the sulfate and nitrate ions are predominantly anthropogenic in origin, having been formed in the atmosphere from NO and SO_2 precursors emitted as byproducts of the combustion of fossil fuels.

Because acid rain is found in rural areas downwind of industrialized regions, it is clear that precursors are transported in the atmosphere for long distances, of the order of hundreds of kilometers, before being deposited in wet form. During this period of travel, which may require a day or more, the precursors are oxidized to the acidic ions, SO_4^{2-} and NO_3^-, via complex processes which are not well understood at the present time. The precise relationship between the amount of precursor emissions and the rate of wet acid deposition at remote distances from the emission sources is not known unequivocally because of the complexity of the intervening processes. It is the objective of long range transport models to provide quantitative information on such source-receptor relationships for use in acid rain control programs.

Models which describe the transport, transformation and deposition of air pollutants at long distances from the source (of the order of 10^3 km) are of two types: Lagrangian and Eulerian. The former utilize air parcel

trajectories computed from historical meteorological data to relate source emissions to receptor effects, while the latter describe average flow properties within an Eulerian grid. This paper is concerned with an Eulerian model averaged over a long time period (several months to a year) and utilizing the statistical properties of air parcel trajectories. Such a model has been termed a climatic model to distinguish it from episodic models which attempt to describe the temporal changes in air pollution over a period of several days within a large region but containing fine spacial resolution. Climatic models show less spatial and time resolution than do episodic models.

The transport and transformation of acid rain precursors occurs within a mixed layer at the base of the atmosphere whose height is of the order of a few kilometers. But because the transport occurs over hundreds of kilometers, the dispersion of the pollutants is primarily in the two lateral dimensions parallel to the earth's surface. While the most elaborate models include vertical mixing within the mixed layer as an element, most models, like ours, assume that the primary and secondary pollutants are well mixed vertically and hence the variables of interest, such as concentrations of airborne species and deposition rates, are functions of latitude and longitude only.

A critical element in any model is the specification of the rate processes for chemical transformation of primary precursors to secondary acidic species and deposition of either species to the earth's surface. These complex processes are represented by first order linear terms in the mass conservation equation for each species. While this cannot be precisely true, especially for the transformation processes, it may be a

sufficiently good approximation to the actual process to preserve the usefulness of a model for analytical or predictive purposes.

The long distance transport model discussed below was devised by Fay and Rosenzweig (1) to describe the long distance transport of primary and secondary pollutants. It provides for the conversion of primary to secondary pollutant species and the wet and dry deposition of both species as linear processes. By virtue of the linear approximation, the contributions from various sources can be added together, greatly augmenting the usefulness of the model. In this paper we extend and simplify the model for the purpose of evaluating acid rain control programs.

DISPERSION MODEL

The long distance dispersion model of Fay and Rosenzweig (1) assumes that the horizontal dispersion can be expressed by a turbulent transport model having a horizontal eddy diffusivity D which is constant throughout the flow field. Averaging over long times and assuming that the mean wind speed w is also constant in magnitude and direction everywhere, the conservation of mass of primary pollutant, whose mass concentration is X_p, becomes

$$w\partial X_p/\partial x = D(\partial^2 X_p/\partial x^2 + \partial^2 X_p/\partial y^2) - X_p/\tau \qquad (1)$$

in which x and y are the streamwise and normal distances and τ^{-1} is the rate constant for loss of primary pollutant due to wet and dry deposition and conversion to secondary pollutant.

We next find a solution to Eq. (1) for a source of mass flux Q of primary pollutant located at the origin (x=0, y=0):

$$\chi_p = (Q/2\pi hD)\exp(x/\lambda)K_o\{r/\ell\} \qquad (2)$$

in which h is the height of the mixing layer, r is the radial distance from the source to the receptor, $K_o\{z\}$ is the modified Bessel function of zero order and argument z, and the lengths λ and ℓ are defined by:

$$\lambda \equiv 2D/w \qquad (3)$$

$$\ell^{-2} \equiv \lambda^{-2} + (D\tau)^{-1} \qquad (4)$$

so that $\ell \leq \lambda$. Because the dispersion equation (1) is linear, we may superpose any collection of sources to determine the concentration χ_p at a chosen receptor location. Near the origin, χ_p varies in proportion to $\ell n(r/\ell)$. This is a weak singularity which can be avoided in a practical calculation (1). Eq. (2) breaks down at r = 0 because Eq. (1) assumes perfect vertical diffusion at the source. At very large distances from the source, χ_p decreases exponentially with r, more rapidly in the upwind than in the downwind direction.

A mass conservation equation similar to Eq. (1) can be written for the secondary pollutant concentration χ_s:

$$w\partial\chi_s/\partial x = D(\partial^2\chi_s/\partial x^2 + \partial^2\chi_s/\partial y^2) - \chi_s/\tau_s + \beta\chi_p/\tau_c \qquad (5)$$

in which τ_s^{-1} is the rate constant for loss of secondary pollutant due to both wet and dry deposition, τ_c^{-1} is the rate constant for conversion of primary to secondary pollutant (which must be less than τ^{-1}), and β is the mass of secondary pollutant formed per unit mass of primary pollutant. For a source of primary pollutant at the origin, Eqs. (2) and (5) can be solved to yield:

$$X_s = (\beta Q/2\pi hD)\exp(x/\lambda)[K_o\{r/\ell\} - K_o\{r/\eta\}]/[(\zeta/\eta)2 - (\zeta/\ell)^2] \quad (6)$$

in which the lengths ζ and η are defined by:

$$\zeta^2 \equiv D\tau_c \quad (7)$$

$$\eta\text{-}2 \equiv \lambda^{-2} + (D\tau_s)^{-1} \quad (8)$$

Note that both η and ℓ are less than λ, but ζ may be more or less than λ.

Fay and Rosenzweig (1) suggest the following values for the dimensional parameters of Eqs. (2) and (6): $w = 2ms^{-1}$; $D = 2x10^6m^2s^{-1}$; $\tau = 8x10^4s$; $\tau_s = 1.6 \times 10^5s$; $\tau_c = 2.4 \times 10^5s$. The corresponding length scales are $\lambda = 2 \times 10^6m$; $\ell = 3.9 \times 10^5m$; $\zeta = 6.9 \times 10^5$; $\eta = 5.4\ 10^5$. Thus the length scales are of the order of 100 km except for λ, which is an order of magnitude larger. As consequence, the factor $\exp(x/\lambda)$ varies more slowly than the Bessel functions in Eqs. (2) and (6), suggesting that the convective term in Eqs. (1) and (5) is not of great importance.

In Fig. 1 is reproduced Fig. 3 of Fay and Rosenzweig showing a comparison of the annual average of the secondary pollutant, airborn sulfate, as calculated (solid line) from Eq. (6) and as measured (dotted line through smoothed values) at monitoring stations indicated by the small circles. This comparison includes an assumed background level of 6 μgm^{-3}. One can see that the agreement is satisfactory over the eastern half of the United States where substantial rain acidity has been observed (2).

It is interesting to determine the fraction of the primary pollutant which is ultimately deposited in the form of wet or dry deposition of

secondary pollutant. Since the rate constant for conversion of primary to secondary pollutant is τ_c^{-1} and the rate constant for deposition of primary pollutant is τ^{-1}, and these rates are the same everywhere throughout the flowfield, the ratio τ/τ_c must be the ratio of secondary to primary deposition rates. For the values of τ and τ_c quoted above for SO_2 emissions, τ/τ_c is 1/3. This may be compared with Golomb's (3) estimate that 0.18 of SO_2 emissions in the eastern U.S. is wet deposited as sulfate. Galloway and Whelpdale (2) estimate that dry deposition is about the same as wet deposition. Thus the quoted value of 1/3 for τ/τ_c is compatible with these estimates.

It is possible to simplify the form of Eq. (6) if $|r/\ell - r/\eta| \ll 1$ or $r \ll |\ell^{-1} + \eta^{-1}|^{-1}$. Using the values of ℓ and η quoted above, this would require that $r \ll 1.4 \times 10^6$m. But at distances as great or greater than this limit χ_s would be very much smaller than its value near the primary source, and hence of little practical interest. In this limit Eq. (6) takes the form:

$$\chi_s = (\beta Q/4\pi hD)(s/\zeta)^2 \, [\exp(x/\lambda)] \, (r/s) \, K_1(r/s) \tag{9}$$

in which K_1 is the modified Bessel function of order one and the length s is:

$$s \equiv (\ell + \eta)/2 \tag{10}$$

and has a value of 4.7×10^5 m if the values of ℓ and η are those quoted above. If $\lambda \gg s$ the exponential factor in Eq. (9) can be neglected without much loss of accuracy in determining χ_s.

The distance-dependent factors (the last two) of Eq. (9) depend only upon the normalized distance r/s. Fig. 2 shows how these factors

decrease as a function of r/s, ultimately decaying exponentially according to the asymptotic limit:

$$(r/s)\ K_1\{r/s\} \simeq (\pi s/2r)^{1/2}\ \exp\{-r/s\} \qquad (11)$$

Note, however, that the secondary pollutant concentration is nearly uniform near the primary source (r=o) and does not substantially decay until r/s exceeds unity.

Of particular interest is the concentration χ_{so} of secondary pollutant at the source (r = o) of the primary pollutant. In this limit Eqs. (6) and (9) reduce to, respectively,

$$\chi_{so} = (\beta Q/2\pi hD)\ \ell n\ (\ell/\eta)/[(\zeta/\eta)^2 - (\zeta/\ell)^2] \qquad (12)$$

$$= (\beta Q/2\pi hD)(s^2/2\zeta^2) \qquad (13)$$

where Eq. (11) is the equivalent of Eq. (12) of Fay and Rosenzweig (1).

SOURCE APPORTIONMENT

Source apportionment is a methodology for determining the fraction of the concentration χ which is due to each source i (of strength Q_i) among a collection of contributing sources. Our linear model may be used to determine the fractions f_{si} of secondary pollutant which are caused by the primary sources Q_i. First we determine the transfer coefficients k_{si} relating the receptor concentration to the source strength:

$$k_{si} \equiv \chi_s/Q_i \qquad (14)$$

using Eq. (6) or (9) to evaluate χ_s/Q_i. Then the source apportionment fraction f_{si} is found from:

$$f_{si} = k_{si}\ Q_i/\Sigma_i\ k_{si}\ Q_i \qquad (15)$$

Note that the fractions f_{si} are independent of the absolute value of X_s as determined by the leading factor in Eq. (6) or (9) and depend only upon the distance related factors (as illustrated in Fig. 2) and, of course, the primary source strengths Q_i.

To illustrate the usefulness of the source apportionment fractions, we show in the first column of Table 1 the percentage of annual average ambient sulphate at a receptor located in the Adirondack mountains as calculated from Eqs. (14) and (15) of this paper. The sulfur sources considered are those in a 31 state region bordering on and east of the Mississippi River (4). Ten states or groups of states (14 states all told) are listed in rank order of their contribution to the Adirondack sulfate level. Their cumulative contribution is more than 80% of the total from the 31 states. It can be seen from the list that the major contributors are the nearby states in the northeast region of the U.S. Nearly half of all the ambient sulfate from the 31-state group is caused by the states which share a border with New York (including, of course, New York itself).

Table 1 also shows a comparison with similar calculations based on results from models which were reviewed by the U.S. - Canada Regional Modeling Subgroup (5). Of these models, MOE is a climatic model similar to that of this paper; ASTRAP, ENAMAP and UMACID are trajectory models; MEP and AES are Lagrargian models and CAPITA is a Monte Carlo model. Despite the quite different features of these models, the principal results are quite similar; i.e., the predominant effect of nearby sources and the negligible contributions of the more distant ones. The implications of these source apportionment analyses for acid rain control strategies are discussed by Fay and Golomb (5).

The last column of Table 1 lists the geometric mean of all eight entries.

CONCLUSION

The climatic model of dispersion of acid rain precursors described in this paper is an easy one to apply to the determination of the relative contributions of various sources to the ambient concentrations of secondary pollutants. Like most other linear models, it makes use of the principle of superposition to determine source-receptor relationships.

The relationship of secondary pollutant level to primary source strength, distance and orientation to the average wind involves the use of several parameters characterizing the importance physical and chemical processes. Assumed values of these parameters determine the quantitative results but the qualitative features of the model are related to the model assumptions regarding the transport, transformation and deposition of pollutants.

An example of the determination of the source apportionment fractions of air-born sulfate at an Adirondack receptor shows the dominant influence of nearby sources compared with distant sources. These features are quantitatively similar to those derived from seven other models of greater complexity.

ACKNOWLEDGMENT

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TABLE 1

Comparison of Model Predictions for % of Ambient Sulphate at Adirondack Receptor+

Source State \ Model	This paper	MOE*	ASTRAP*	ENAMAP*	UMACID*	MEP*	AES*	CAPITA*	Mean and S.D.
PA	17.4	13.2	18.1	19.7	15.9	23.6	19.5	18.6	18.3 ± 3.0
OH	14.8	16.2	15.9	26.7	20.9	17.4	18.8	17.7	18.6 ± 3.8
NY	10.5	9.4	24.2	9.3	12.4	23.4	13.3	10.1	14.1 ± 6.2
IN	7.6	9.5	7.8	9.7	8.1	5.1	7.2	8.9	8.0 ± 1.5
WV	6.6	5.2	4.9	7.9	5.7	5.6	6.7	8.9	6.4 ± 1.4
MD/DE/NJ/DC	6.4	3.4	4.2	2.9	3.5	5.5	4.0	3.1	4.1 ± 1.2
MA/CT/RI	5.8	2.9	1.3	2.7	4.7	3.6	2.6	1.7	3.2 ± 1.5
MI	5.2	7.8	3.9	7.3	7.2	6.1	9.0	5.9	6.6 ± 1.6
IL	3.9	6.5	4.0	2.7	5.1	2.3	6.0	5.4	4.5 ± 1.5
KY	3.4	3.5	3.7	4.4	3.0	1.7	2.1	3.8	3.2 ± 0.9
Other	18.4	32.4	12.0	6.7	13.5	5.5	10.8	15.9	14.4 ± 8.5

+ Sources in 31 state region only (4)

* Data from (5)

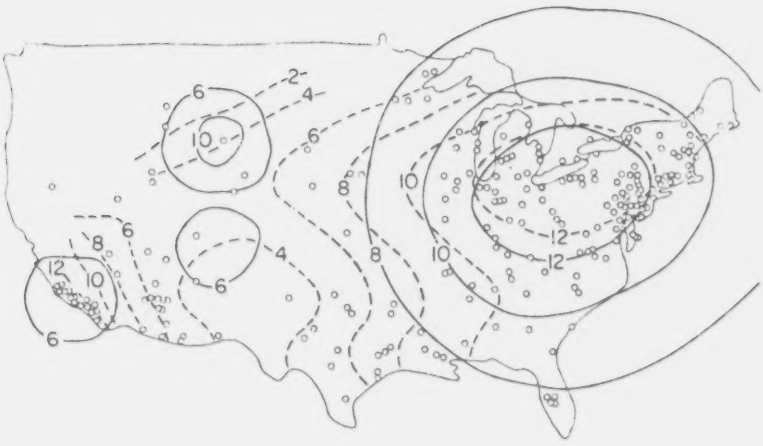

Fig. 1 A comparison of the annual average sulfate concentration ($\mu g\ m^{-3}$) in the U.S. as calculated (solid line) by Fay and Rosenzweig (1) and as measured (dotted line through smoothed values) at monitoring stations indicated by small circles. From (1).

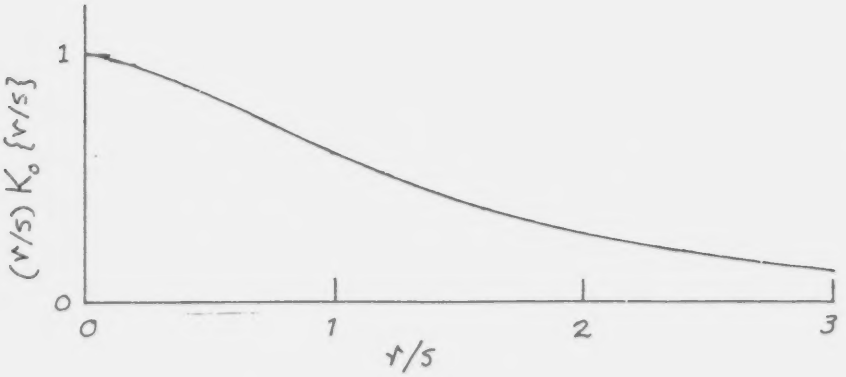

Fig. 2 Space dependent factor of secondary pollutant concentration (Eq. 9).

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CONTROLLING ACID RAIN

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Energy Laboratory Report No. MIT-EL 83-004

Summary. High concentrations of sulfuric and nitric acid in rain in the northeastern USA are caused by the large scale combustion of fossil fuels within this region. Average precipitation acidity is pH 4.2, but spatial and temporal fluctuations of ±1 pH unit have been observed. Amelioration of rain acidity requires significant reduction of precursor emissions--the oxides of sulfur and nitrogen. Such reduction of emissions from existing sources will be difficult and expensive. A pending legislative proposal to reduce eastern U.S. emissions of SO_2 by 10 Mty^{-1} below the 1980 level of 22.5 Mty^{-1} would reduce the acid sulfate deposition rate in the Adirondack mountains, an environmentally sensitive area, from 30 to 21.4 kg $ha^{-1}y^{-1}$. Based upon source/receptor modeling, an equal reduction of Adirondack sulfate deposition rate could be achieved by a 7 Mty^{-1} SO_2 emission reduction if the reduction is allocated according to source proximity to the sensitive area. The cost of reducing 10 Mty^{-1} SO_2 emissions is estimated at $5 to $8 billion per year. Considerably lower costs could be realized in an emission control scheme that is the more stringent the nearer the sources are to the environmentally sensitive areas, and if novel approaches are implemented, such as seasonal or episodic emission reduction, NO_x vs. SO_2 emission control, emission redistribution, and least emission electricity dispatch. Substantial emission reductions will probably be achieved in the more distant future by employing new combustion technology, such as lime injected multistage combustion, fluidized bed combustion, coal gasifier combined cycle, magnetohydrodynamics, and possibly others.

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Overview

Acid rain is a severe environmental problem, one that is difficult to ameliorate. The origins of precipitation acidity are well known--the oxides of sulfur and nitrogen formed in the combustion of fossil fuels. By processes which are understood only in general but not in particular, these emissions are carried great distances, being transformed during passage to acidic species and then deposited in wet or dry form on the surface of the earth. Millions of tons of acidic material are thus deposited in eastern North America each year, some of it in areas which are so ecologically sensitive as to suffer damage to aquatic systems or forest crops. The most direct attack on the acid rain problem involves reducing significantly emissions of acid precursors--the oxides of sulfur and nitrogen--by modifying the fuel or combustion process at the source. Complete elimination of such emissions is out of the question and substantial reductions, while technologically feasible, will be very expensive in the aggregate.

The ecological damage from acid rainfall is not uniform throughout this region. Principally mountainous regions of eastern U.S. and Canada, called sensitive areas, are affected because of their limited capacity to buffer acid precipitation. Aquatic systems show most clearly such effects, but forest species appear not to be exempt from damage. On the other hand, crops grown in predominantly agricultural regions seem not be susceptible.

To provide some environmental relief to these sensitive areas, Congress is considering the imposition of sulfur oxide emission control plans within 31 states east of or bordering on the Mississippi River.

The more ambitious proposal, the Mitchell bill, would reduce annual SO_2 emissions by 10 million tons, that is, by about 45 percent of the present emission level. It is estimated that the effect of such an emission reduction on the average precipitation acidity would be an increase of pH (i.e., reduction of acidity) by 0.1-0.2 units above the current mean value of 4.2. However, the annual deposition rate of sulfate will decrease commensurately to the emission reduction of SO_2. Whether environmental benefits are related to changes in pH or reduction of sulfate deposition rates, many sensitive areas would not be removed from the endangered list by this level of amelioration. Further reduction of SO_2 and NO_x emissions would be required to reduce precipitation acidity to harmless levels.

While acid species are carried long distances from their precursor sources, a source closer to a sensitive area causes greater deposition than an equal source much further away. Such source-receptor relationships, although presently not precisely defined, provide a basis for considering advanced control programs for SO_2 and NO_x emissions that would be more cost-effective. Such alternative strategies would concentrate the emission reductions among those sources closest to the sensitive areas. These cost-beneficial strategies become especially important for future reductions beyond those already contemplated, which may be more expensive and will undoubtedly involve balancing the relative costs and benefits of SO_2 *vs.* NO_x controls.

Even within the proposed control programs, but especially for more comprehensive ones, there are alternatives to the allocation of emission reductions which could decrease deposition rates or the cost of control. Interchange of high and low sulfur fuels, electric power generation and

transmission between regions close to and far from sensitive areas, least emission dispatch of electric power, and seasonal (summer) control of emissions have the potential for increasing the cost-effectiveness of control. Also, a different balance of SO_2 and NO_x reductions among such regions might be beneficial. The greater the degree of emissions reduction which is desired, the higher are the marginal costs of reduction and the more varied are the methods needed and sources to be controlled.

The available methods for reducing sulfur emissions from existing sources consist principally of (1) removing sulfur from the fuel before combustion, (2) the removal of sulfur dioxide from flue gases, and (3) substitution of low sulfur for high sulfur fuel. But new technology under development would remove sulfur during the combustion process or in a precombustion process, hopefully more cheaply and effectively. For reducing NO_x emissions the principal method is combustion modification. Presently practiced methods can be applied to existing plants by retrofitting, but developing technologies require new, compatible boiler design, and even completely new plants which may be located elsewhere than the existing ones.

Gearing up for acid rain mitigation programs will require many years and enormous investments in supplying emission control devices and/or low sulfur fuels. In the meantime, to offset the deterioration of the most threatened aquatic systems a program of liming of lakes, and possibly forested areas, may be needed. But there is little expectation that such palliatives, by no means inexpensive, can offset indefinitely present levels of acid deposition.

The Sources and Effects of Acid Rain

Precipitation acidity. While it has been known for more than a hundred years that rainfall sometimes is quite acidic, only within the past few decades has there arisen a recognition that acid rainfall deleteriously affects natural ecosystems, especially fresh water fish populations. Since World War II, first in Scandinavia and then in northeastern North America, monitoring of rainfall acidity and aquatic ecosystems has shown some evidence of increasingly severe aggregate effects. The acidity is principally associated with sulfate and nitrate ions in precipitation, although other, less prominent species may add to or subtract from the overall acidity. These principal ions are formed from the oxidation of sulfur and nitrogen oxides (SO_x and NO_x) in the atmosphere whose principal (but not sole) sources are the products of combustion of fossil fuels.

Part of the emitted gaseous SO_2 and NO is oxidized to form the SO_4^{2-} and NO_3^- ions which are deposited in raindrops. This process is thought to be quite complex, involving several pathways of gas, liquid and solid phase oxidation. The presence of water vapor, oxidants such as OH and O_3, and sunlight are considered to be essential in this transformation of primary to secondary air pollutants. After many hours and even days, the acid sulfates and nitrates so formed exist in the atmosphere as chemical components of very fine particulate matter, including liquid droplets. Some of this material can be incorporated into raindrops as they are formed or is scavenged from the air by rainfall originating at higher altitudes. Because these atmospheric processes are very slow, the precipitation of sulfate and nitrate ions

occurs to great distances from the point at which combustion effluent is injected into the atmosphere.

The acid content of rain is measured by its hydrogen ion concentration. This concentration is defined on a logarithmic scale, each unit decrement in pH corresponding to a tenfold increase of hydrogen ion concentration. Pure water has a pH of 7. Rain water in equilibrium with the atmospheric carbon dioxide has a pH value of 5.6. A pH of 4 could be formed by one part per million of sulfuric acid or 2 ppm of nitric acid (or any proportionate combination of both). Typical rainfall pH, during episodes of noticeable acidity, is in the range of 3.5 to 4.5. Hence acid concentrations in rain, on average, are about one part per million. On the other hand, the concentration of NO_x and SO_x in the atmosphere through which the rain descends is about ten parts per billion. Because of chemical and physical factors, precipitation scavenges and concentrates the airborne species, delivering an acidic solution to the surface of the earth.

It is a common observation that rainfall is an episodic phenomenon, i.e., precipitation occurs during a small percentage of the time. The acidity of rainfall varies greatly from one rainfall event to the next, and even during a single rainfall. Peak hydrogen ion concentrations can be ten times higher than the annual average values. Thus much of the annual acidity deposited by rain may be contained in only a small fraction of the total rainfall which falls during a very small percentage of the time.

Acidic rainfall is most noticeable near regions of heavy industrial activity, such as western Europe and eastern North America. Within such identifiable areas, estimates of sulfur sources show a ten-to-one

preponderance of man-made emissions over biogenic sources. In the U.S., about 90 percent of anthropogenic sulfur is released in the combustion of fossil fuels, about 2/3 of this by electric generating stations (Table 1). Of the total sulfur released into the atmosphere within the eastern U.S., about 20 percent ends up in acid precipitation and 80 percent in dry deposition and convective transport by the wind beyond the region's boundaries (1). (Of course, all of the emitted sulfur ultimately returns to the surface of the earth or ocean.)

Table 1. Distribution of U.S. SO_2 and NO_x emissions by source category. From (2).

Category	SO_2 (percent of total)	NO_x (percent of total)
Utilities	65	31
Industrial boilers	12	21
Industrial processes	15	3.5
Transportation	3	40
Residential/commercial	5	3.5
Solid waste disposal	0	0.5
Miscellaneous	0	0.5
Total	100	100

A similar proportioning of sources and sinks also holds for nitrogen oxides. Of the combustion sources of NO_x, vehicles (principally the automobile) account for nearly half. In terms of acidifying potential, aggregate NO_x emissions constitute about one-third of the total and SO_2 the remaining two-thirds. However, these proportions will be

different at the point of acid precipitation since the source streams are depleted at different rates.

Over the past few years, high quality precipitation chemistry monitoring stations were established across North America. These stations measure precipitation acidity (hydrogen ion concentration) and other important cations and anions in rain, snow, dew, etc. Fig. 1 shows the data obtained in 1980 for hydrogen ion deposition rates (2). The units are millimoles $m^{-2}y^{-1}$, obtained by multiplying the mean concentrations in precipitation by the amount of precipitation at the site. The discrete numbers are the records from individual stations; the contours (isopleths) are hand-drawn and therefore somewhat subjective. From measurements of this type it has been found that hydrogen ion deposition reaches a maximum in the Ohio-Pennsylvania-West Virginia triangle, but depositions in excess of 50 mmoles $m^{-2}y^{-1}$ (equivalent to an annual average pH of 4.3, assuming 100 cm y^{-1} precipitation) occur as far west as Illinois, north to southern Ontario and Quebec, east to the Atlantic seabord, and south to North Carolina and Tennessee.

Relating emissions to deposition. Quantitative estimates of the relation between emissions of acid rain precursors and amount of acid deposition are based upon mathematical models of the physical and chemical processes of atmospheric transport, transformation and deposition. There is a great variety of such models--a recent study enumerates 43 of them--some of which are very complex. As the detail of description in time and space generated by a model increases, so does its complexity, cost of computation and the degree of detailed specification of the meteorological, chemical and physical processes believed to be at work. Current studies of model development seek to improve the

Fig. 1. Precipitation-weighted hydrogen ion deposition in eastern North America (mmoles $m^{-2}y^{-1}$). Crosses mark box area discussed later. From (2).

reliability of models through modifications which increase the correlation between measured and calculated variables, such as acid deposition rates, etc.

A nearly universal and simplifying assumption of acid rain modeling is that of linearity, that is, all physical and chemical processes (e.g., dry deposition, oxidation of precursors) proceed at a rate proportional to the concentration of the pollutant in question. When this is so, the effects of pollutants emitted from each source may be added together at each receptor so that the apportionment of the effects produced at the receptor may be allocated quantitatively to the various sources, no matter how distant. But where processes are not linear, the linearity hypothesis will only approximate the true rates and the resulting conclusions regarding acid deposition may not be accurate. Only further research of the physicochemical processes and nonlinear modeling can resolve whatever uncertainties now exist regarding the linear hypothesis.

The simplest model is the chemical mass balance or box model. For example, consider the atmosphere which covers the U.S. east of the Mississippi as a box into which SO_2 and NO_x emissions from all sources are introduced at a constant average rate. Since the mass of elemental sulfur and nitrogen is conserved, these species are lost at an equal rate from the box because of wet and dry deposition to the earth's surface or by net advection by the wind out of the boundaries of the box. Using atmospheric measurements one can estimate the various components of this mass balance and thus determine in what form and proportions the emissions leave the box. The box model provides the simplest argument which relates acid rain deposition to anthropogenic emissions of SO_2 and NO_x.

Neither acid rainfall nor precursor emissions are uniformly distributed throughout such a large region. To obtain a more detailed understanding of the mass balance, the region can be subdivided into a large number of microscopic box models--say 50 x 50 km in base area. By performing simultaneous mass balances in all of these small elements, a much more detailed description of acid deposition within a broad region can be obtained. But because the relevant atmospheric monitoring has been carried out in only a few of these microscopic boxes, the atmospheric processes of transport, transformation and deposition must be modeled mathematically in each box in order to accomplish an overall mass balance. Since knowledge of these processes is only approximate, uncertainties about the model predictions are thereby introduced.

Episodic models seek to describe the daily changes of precursor behavior and acid rainfall through a detailed tracking of the meteorological events which govern the fate of pollutant between source and receptor. Because of the great expense of such modeling, attention is first focussed on the explanation of high pollutant episodes lasting several days to test the performance of such models.

One method of reducing the complexity of acid rain modeling is to average the mass balance over a long time period--say a month, season or year. The resulting models are called climatic or statistical models. The climatic models can provide average acid deposition rates without requiring detailed information of meteorological events on a diurnal time scale. Time-averaged deposition may prove to be an acceptable surrogate for evaluating environmental effects and the corresponding benefits from mitigation strategies.

Identifying areas sensitive to acid rain. Fresh water aquatic

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ecosystems are adversely affected by the acidification of pond or lake water, but in ways which are also determined by the kind and proportions of anions and cations rather than simply by the net hydrogen ion concentration. In many cases precipitation enters a lake principally via a drainage basin where the acidic composition may be altered. Where it exists, the buffering capacity of the drainage basin and lake bed can substantially modify the acidic quality of the rainfall and prevent the increase of lake or pond acidity. Given the complex chemical processes at work in aquatic ecosystems, it is difficult to predict in detail how they would respond to changes of precipitation acidity.

Many lakes in the northeastern U.S. and eastern Canada have been markedly affected by acid precipitation because of the lack of natural buffering capacity, the underlying geologic structure being granite rather than limestone. Because acid rainfall is widespread, sensitive receptor areas can be identified by surveying for the early signs of ecosystem deterioration among the aquatic ecosystem populations. It is these sensitive areas which would be the principal beneficiaries of reductions of acid precipitation.

The damage to forests from acid rain is less certain and quantifiable. Soil characteristics and tree species will determine the effects. Given the ubiquity of silviculture in the northeastern and southeastern U.S., even minor damage caused by acid rain could aggregate to significant economic impacts.

Galloway and Cowling (3) defined the acid-susceptible areas of North America in terms of surface waters contained in igneous and metamorphic rock. Their sensitivity map is reproduced in Fig. 2. Omernik and Powers (4) developed a new sensitivity map based on total alkalinity of surface

Fig. 2. Regions of North America (shaded) containing lakes sensitive to acidification. From (3).

waters. This map permits a better resolution for identifying watersheds that are susceptible to acidification than is shown in Fig. 2.

The Impact Assessment Interim Report produced under the auspices of the United States-Canada Memorandum of Intent on Transboundary Air Pollution (5) identified the following watersheds that might be sensitive to acidic deposition:

- o Northern Michigan, Wisconsin, Minnesota
- o New England
- o Adirondack region of New York
- o Southeastern New Jersey
- o Western North Carolina and parts of the Smoky Mountains
- o Southeastern Ontario
- o Southeastern Quebec
- o Maritime Provinces and Newfoundland

The U.S./Canada report cautions that "a more comprehensive and verifiable estimate of acuatic acidification carrying capacity must await the findings of research presently being conducted."

In addition to identifying areas sensitive to acid deposition, it is important to relate them to present deposition patterns. Obviously, only sensitive areas that are exposed to excessive acid loading will become acidified. However, there is no unambiguous criterion of what constitutes an excessive loading and it is not even clear whether sulfate, nitrate or hydrogen ion loading (or a combination of these ions) causes the ecological damage. Recent assessments (5,6,7) indicate that sulfate deposition may be a good indicator for environmental damage. They suggest that at a sulfate loading of 15 kg ha^{-1} y^{-1}, only the most sensitive lakes become acidified; at a loading of 45 kg ha^{-1}

y^{-1}, moderately sensitive lakes also become acidified, and sensitive species become extinct. Using a 30 kg $ha^{-1}y^{-1}$ threshold, the exposed sensitive areas are confined to upper New York state, New England, southern Ontario and Quebec and parts of the Smoky Mountains.

Emission Reduction/Acid Deposition Relationship

Box model. Simple acid rain models may be used to estimate the effects on acid deposition of changes in emission rates of the precursors SO_2 and NO_x. The commonly used linear model predicts that the acid deposition effects of many sources is simply the sum of the effects from each source considered separately. The simplest of linear models is the mass-balance-in-a-box model described by Golomb (1) and summarized here.

Let us take a box of sufficient size such that most of the transport and transformation processes occurs within the box. We account for all sources in the box: emissions and influx from outside the box, and all sinks: deposition (wet and dry) and efflux through the walls. The "sufficient" size stipulation is only necessary to ensure that the influx is small compared to the emissions.

The northeast quadrant of the U.S. was selected as a suitable box. The boundaries run from the SW corner of Tennessee to the NW corner of Illinois, east to the NE corner of New Hampshire, south to the SE corner of North Carolina, and back to Tennessee. The box is marked with crosses in Fig. 1. The bottom area of the box is 1.2×10^6 km^2. For a steady state to be established, a sufficient time period needs to be chosen, say one year. The year 1978/79 was analyzed. The emissions within the box are 2.2×10^{11} moles y^{-1} of SO_2 and 1.8×10^{11}

moles y^{-1} of NO_x, with an estimated uncertainty of ±10 percent.

The wet-deposition of sulfate, nitrate and other ions inside the box was analyzed by Pack (8) based on the MAP3S and SURE monitoring network data. Integrating the Pack isopleths (which are similar to Fig. 1), the total annual wet-deposition of sulfate amounts to 4.3×10^{10} moles y^{-1}, and nitrate 3.9×10^{10} moles y^{-1} (with an uncertainty of ±25 percent).

Thus, it is found that 19 percent of the emitted SO_2 is wet-deposited as sulfate within the box, and 20 percent of the emitted NO_x is wet-deposited as nitrate. The rest evidently follows other pathways such as dry deposition and net efflux. Similar results for eastern North America were obtained by Galloway and Whelpdale (9), and for western Europe by Garland (10) and Rhode (11). The average concentration in precipitation is 27 μmoles l^{-1} of sulfate and 24 μmoles l^{-1} of nitrate. If every sulfate ion carried two hydrogen ions and every nitrate one, the average H^+ concentration would be 78 μmoles l^{-1}, pH = 4.1. However, some neutralization occurs due to the presence of ammonium, calcium and magnesium ions and the corrected average H^+ concentration is 67 μmoles l^{-1}, pH = 4.2 (1). This is in good agreement with the mean of observations (12). Note that the calculated values are averages; local and temporal fluctuations by a factor of 10 in hydrogen ion concentrations (pH ±1.0) have been observed (12). Sulfate/nitrate ratios are also subject to spatial and temporal variations. However, on the average, 2/3 of the precipitation acidity is due to sulfuric acid and 1/3 to nitric acid.

Proportionate emission reduction. The simplest case to consider is an across-the-board reduction of SO_2 and NO_x. If both emissions and

depositions are averaged over a large geographic area, such as the eastern U.S., then the spatially averaged deposition rates of each species would be proportional to the aggregate emission rates, assuming that the latter are reduced in a geographically uniform way. For example, a tenfold reduction of emissions would result in a tenfold decrease of wet-deposited ions everywhere within the zone of influence of the sources whose emissions are reduced. Of course, across-the-board reductions are impractical to achieve, especially for NO_x, and certainly not tenfold.

Table 2 lists the effect of proportionate emission reduction on the annual average rain acidity by successive elimination of the source categories of Table 1. From Table 2 it becomes evident that significant improvement of the average acidity can only be achieved by very substantial curtailment of emissions. Since such area-wide curtailments may not be technically and economically feasible, a more effective approach, at least in the near term, would be the reduction of emissions primarily in source areas that can be shown to contribute most of the acid deposition to selected sensitive areas.

Source apportionment modeling. To provide alternative approaches, it is necessary to resort to a particular model which takes into account the fact that a more distant source has a lesser effect than a nearby source on the amount of acid deposition, other things being equal. Using the climatic model of Fay and Rosenzweig (13), Table 3 lists the percent contribution to acid sulfate in rainfall at a receptor in the Adirondack Mountains caused by the aggregate SO_2 emissions in each of the states in the eastern U.S., by rank order of their respective contributions.

Table 2. Expected mean regional precipitation acidity resulting from elimination of emissions from source categories

Case	Emission reduction (percent below present)		Precipitation acidity*	
	SO_2	NO_x	$[H^+]$ μ moles l^{-1}	pH
(1)	0	0	67.0	4.2**
(2)	65	0	38.2	4.4
(3)	65	31	31.1	4.5
(4)	77	0	32.9	4.5
(5)	77	52	21.1	4.7
(6)	92	95.5	7.0	5.15
(7)	100	100	2.5	5.6

1. The present estimated and observed average acidity in the northeastern U.S.. 2. Elimination of SO_2 emissions from all power plants. 3. Elimination of SO_2 and NO_x emissions from power plants. 4. Elimination of SO_2 emissions from power plants and industrial boilers. 5. Elimination of SO_2 and NO_x emissions from power plants and industrial boilers. 6. Elimination of emissions from all major industrial sectors and transportation. 7. No emissions whatsoever, the acidity of water in equilibrium with CO_2 (carbonic acid).

*Probable error bound ±30 percent of $[H^+]$, ±0.15 pH.

**Large local and temporal fluctuations have been observed; factor of 10 in $[H^+]$, ±1.0 pH.

Table 3. Relative contributions of U.S. sources to acid sulfate in rainfall at Adirondack receptor

State	Contribution (percent of total)	Rank by source strength
Pennsylvania	17.4	2
Ohio	14.8	1
New York	10.5	9
Indiana	7.6	3
West Virginia	6.6	7
Michigan	5.2	11
Massachusetts	4.1	16
Illinois	3.9	4
Kentucky	3.4	6
New Jersey	2.9	17
Other eastern U.S. states	23.6	
Total*	100	

*The observed annual average acid sulfate deposition at Whiteface Mountain in the Adirondacks in recent years is about 30 kg $ha^{-1}y^{-1}$ (12). Of this, U.S. sources contribute about 70 percent, Canadian sources about 30 percent (5).

(Fay and Rosenzweig's model allows the estimate of ambient sulfate concentrations; in these calculations we assume that sulfate wet-deposition is proportional to ambient sulfate concentrations.) The ten largest contributors aggregate to more than three quarters of the total effect. Also shown is the rank order of the source strengths of each state. Obviously some of the nearby states are greater contributors than would be expected by their source strengths.

The source/receptor relationships listed in Table 3 are those for a particular model. Other models give similar but not identical results. It is hoped that further development of models will provide more reliable estimates of the apportionment of acid deposition at a receptor among the various sources of precursor emissions.

Of particular interest is the relative contribution of U.S. and Canadian sources to acid deposition in the northeastern U.S. and eastern Canada. The U.S.-Canada joint study (5) estimates that more than 30 percent of acid sulfate deposition at an Adirondack receptor originates in Canada. In another study by Galloway and Whelpdale (9), the fractions of domestic emissions which are "exported" to the neighboring country were found to be 14 percent and 33 percent for the eastern U.S. and Canada, respectively. But because Canadian direct emissions are only 15 percent of U.S. emissions, the flow into Canada exceeds that in the reverse direction by a factor of three. However, there is no estimate yet of the percent of Canadian acid rainfall which is due to U.S. sources, and vice versa. Thus the amount of acid deposition which might be mitigated by emissions reduction in both countries is as yet uncertain.

Pending legislative proposals and alternatives. Congress has been considering two major bills for reducing sulfur emissions so as to

mitigate rain acidity. Both propose substantial reductions within a 31 state region east of or bordering on the Mississippi River to be accomplished within ten years. Reduction allocations among the states are based upon 1980 emissions from utility plants. The Mitchell bill would require a reduction of SO_2 emissions of 10 million tons per year, while the Moynihan bill would result in a reduction of 8 Mty^{-1} to be accomplished within 10 years after the enactment. Since the 1980 emissions in this 31 state region are estimated at 22.5 million tons per year, these are reductions of 45 percent and 36 percent respectively. The formula for allocation of these aggregate amounts among the states is different for each bill, but places the greater burden on states which emit, on average, sulfur oxides in excess of 1.2 pounds of SO_2 per million Btu of fuel heating value. Most of the abatement penalty will fall on states which use extensively high sulfur coal in utility boilers. These reductions are presumed to be achievable by substituting low sulfur fuel for currently used high sulfur coal or by desulfurization of flue gases from large sources, principally electric utility plants.

Table 4 compares the distribution of 1980 sulfur emissions among the states, arranged in rank order, with the distribution of reductions required by the Mitchell and Moynihan bills (14). The ten highest emitting states contribute 68 percent of the total emissions from the 31 state region, but would be required to contribute 79 percent and 91 percent of the reductions, respectively. Thus neither bill requires a uniform percentage reduction of emissions among all of the states, but allocates reductions principally to the major emitters, especially so for the Moynihan bill.

Because the proposed emission reductions are not uniformly

Table 4. Allocation of sulfur emission reduction among states

State	Emission (1980)		Emission reduction	
	(kty^{-1})	(percent of total)	Mitchell (percent below 1980)	Moynihan (percent below 1980)
Ohio	2900	12.5	58	53
Pennsylvania	2165	9.6	10	36
Indiana	2080	9.2	57	56
Illinois	1580	6.8	50	41
Missouri	1350	6.0	68	56
Kentucky	1100	4.9	63	71
West Virginia	1100	4.9	53	51
Tennessee	1085	4.8	62	70
New York	1080	4.8	21	7
Florida	985	4.2	36	29
Other (21)	7275	32.3	29	9
All states	22500	100	45	36

Table 5. Effect of proposed sulfur emission reduction on sulfate deposition at Adirondack receptor

State	Emission	Deposition*		
	1980	1980	Mitchell	Moynihan
	(percent of total)	(percent of total)	(percent of 1980 total)	
Ohio	12.5	14.8	6.3	6.9
Pennsylvania	9.6	17.4	10.4	11.2
Indiana	9.2	7.6	3.2	3.3
Illinois	6.8	3.9	2.0	2.3
Missouri	6.0	2.1	0.7	1.0
Kentucky	4.9	3.4	1.3	1.0
West Virginia	4.9	6.6	3.1	3.2
Tennessee	4.8	2.6	1.0	0.8
New York	4.8	10.5	8.3	9.8
Florida	4.2	0.9	0.6	0.6
Other (21)	32.3	30.2	22.2	27.1
All states	100	100	59.1	67.2
Emissions (percent of 1980)			55.2	63.6

*Excludes Canadian contribution to deposition.

distributed among the sources, sulfate deposition rates may not be reduced in proportion to the aggregate emission reduction. We may use source apportionment models to estimate how the emissions reduction will affect the deposition rates at receptors located in sensitive areas. In Table 5 we show such a calculation for a receptor located in the Adirondacks based upon the model of Fay and Rosenzweig (13). In the first two columns are shown, respectively, the distribution of 1980 sulfur emissions and sulfate depositions caused by these emissions (excluding Canadian contributions). We note that the nearby sources contribute, in proportion to their strength, more than the distant sources--especially New York. In the third and fourth columns we have determined the net deposition rates after applying the emission reduction requirement of the Mitchell and Moynihan bills, respectively. It can be seen that the aggregate decrease of the deposition rate in either case is only a few percentage points above the aggregate reduction of emissions, both normalized to their 1980 values. Thus, in spite of the geographic nonuniformity of emission reduction, the aggregate deposition rate is quite commensurate to the aggregate emission reduction in the 31 states.

The measured sulfate deposition rate at the Adirondacks is about 30 kg $ha^{-1}y^{-1}$ (5). Of this, 21 kg is estimated to be due to U.S. sources, 9 kg to Canadian sources. The Mitchell bill would reduce the U.S. contribution by 41 percent, to 12.4 kg $ha^{-1}y^{-1}$. The average annual rain acidity would be expected to increase from the present pH 4.2 to 4.35.

The allocation of emission reduction under these bills is not as effective as it might be if the distances between source states and target receptor were taken into account. For example, New York and

Tennessee have equal 1980 emissions but Tennessee's contribution to the Adirondack deposition is only 35 percent that of New York. Yet under the Mitchell bill Tennessee must reduce its emissions by three times as much as New York. Since it is four times more effective to reduce emissions in New York than Tennessee, at least as far as Adirondack depositions are concerned, it would be logical and probably more cost-effective to require a greater reduction for New York and a lesser reduction for Tennessee than stipulated by the Mitchell bill. This would decrease the aggregate emission reduction but not increase the Adirondack deposition rate.

To illustrate such an alternative, we have calculated an allocation of emission reduction which would achieve the same Adirondack deposition rate as the Mitchell bill and which would conform to one of two requirements: (1) emissions of any state shall not exceed 1980 levels, and (2) for each state the contribution to Adirondack deposition per million Btu of fuel heating value shall be the same. Very distant states (e.g., Louisiana, Arkansas) will not have to reduce emissions at all. States very close to the Adirondacks will have to reduce emissions more than is required by the Mitchell bill. The second requirement would maintain a constant ratio of environmental cost (deposition rate in sensitive area) to local benefit (fuel heat consumed), an equitable criterion for allocating emissions.

The results of such an alternative allocation are shown in Table 6 and compared to the emission reductions of the Mitchell bill. This alternative exempts 12 distant states from any emission reduction below 1980 levels. Of the 10 largest sources, 6 would have smaller and 4 greater reductions than the Mitchell bill requires. In the aggregate,

Table 6. Alternative allocation of sulfur emission reduction

State	Emission reduction (1) Alternative	Emission reduction (2) Mitchell	increased cost (1) − (2)
	(percent below 1980)		
Ohio	59	58	+ 1
Pennsylvania	62	40	+ 22
Indiana	40	57	− 17
Illinois	0	50	− 50
Missouri	0	68	− 68
Kentucky	39	63	− 24
West Virginia	60	53	+ 7
Tennessee	22	62	− 40
New York	59	21	+ 38
Florida	0	36	− 36
Other states (21)	35	29	+ 6
All states	32	45	

the alternative allocation requires a 32 percent reduction of 1980 emissions while the Mitchell bill specifies a 45 percent reduction. This translates into an aggregate emission reduction in the 31 states of 7 Mty^{-1} vs. the 10 Mty^{-1} of the Mitchell bill, yet the environmental effect in the Adirondacks would be the same.

There are, of course, other sensitive areas which might be affected differently than the Adirondacks. Also, allocation of emission reduction based upon other criteria, such as the cost of reduction, might lead to different relations between emission and deposition reductions than the above. Furthermore, trading of emission reductions between states, as is contemplated by the Mitchell bill, will have some differential effect upon deposition rates. All of these aspects deserve more careful scrutiny.

Redistribution of emissions. Atmospheric models help to identify source regions which contribute the bulk of acidity to ecologically sensitive areas. Conversely, these models also delineate regions whose emissions are unlikely to be conveyed to sensitive areas. It may be asked whether such non-contributing regions could possibly increase their emissions above the present level without materially affecting sensitive areas. In such a fashion, the present aggregate emission level in the U.S. could be kept constant, with some regions experiencing an emission reduction and others an increase.

An emission redistribution could be accomplished by a fuel exchange scheme. Acid rain source regions would import low sulfur coal (or other low emission fuel), and export high sulfur coal to non-source regions. In such an exchange, the socio-economic impact on high sulfur coal producing states would be lessened as their product could find a market.

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Exchanges of coal for liquid fuel should also be considered. Schemes can be envisioned whereby low sulfur coal displaces high sulfur coal in power plants near sensitive areas, high sulfur coal displaces oil and gas in power plants in non-source regions, and the displaced oil and gas is used locally or elsewhere for internal combustion engines and petrochemicals, thereby cutting our dependence on imported oil. It should be noted that plants designed exclusively for combustion of liquid fuels (oil or gas) cannot switch to coal without major reconstruction of the boiler unit. Coal substitution will become attractive when the existing plant nears retirement or when liquid fuels near depletion, whichever comes first.

Emission redistribution could also be achieved by long distance transmission of electricity. High sulfur coal could be burned in non-source region power plants and electricity could be transmitted to source region users. In this scheme, the existing power grid would be used to full capacity as well as possible new high voltage (direct current) long distance transmission lines.

It should be recognized that an emission redistribution scheme is contrary to present air quality management practice under EPA's new source performance standards (NSPS). Substitution of coal for oil categorizes the plant as a new source. Present NSPS regulations require 90 percent sulfur removal from high sulfur coal and 70 percent removal of low sulfur coal; i.e., a scrubber installation is mandatory everywhere. For plants that substitute one kind of coal for another, most state implementation plans (SIP) require an emission limit of 1.2 lb SO_2 per MBtu, ruling out the use of high sulfur coal without scrubbing. An emission redistribution scheme might require non-uniform NSPS and SIP variances tailored to meet acid rain environmental goals.

Seasonal and intermittent emission reduction. The concentration of acidic matter in precipitation and its deposition rate is highly variable over time and space. However, the bulk of sulfate and hydrogen ions appears to be deposited in the spring and summer months (12,15). Fig. 3 shows the monthly wet deposition of ions at Brookhaven, NY, from which we estimate that 53.5 percent of the annual total acid deposition falls in the months June-September and 70 percent in May-October. If a similar distribution occurred in the sensitive areas, the annual acid load could be reduced significantly if low sulfur coal would be substituted for high sulfur coal in the summer months, especially in the source regions. Such seasonal fuel switching would also lessen the adverse socio-economic impact on existing coal mining districts that a year-round substitution would entail.

Intermittent emission control during pollution episodes is another possibility for reducing total acid deposition. Evidence is mounting that acidic matter is deposited most heavily following persistent elevated pollution episodes (PEPE). These episodes occur mainly in summer as a consequence of stagnating anticyclonic systems located over eastern North America. This system causes a recirculation of air over the emitting areas with gradual build-up of pollutant concentrations. The phenomenon is manifested by the appearance of haze which can blanket much of the eastern U.S. and Canada. The episode is usually terminated by passage of a cold front or a possible squall line preceding the cold front. The pollutants are effectively scavenged by the ensuing precipitation or thundershowers. If criteria for predicting these episodes were firmly established, emissions could be curtailed at their onset (PEPE alert) and consequently high acid precipitation episodes as

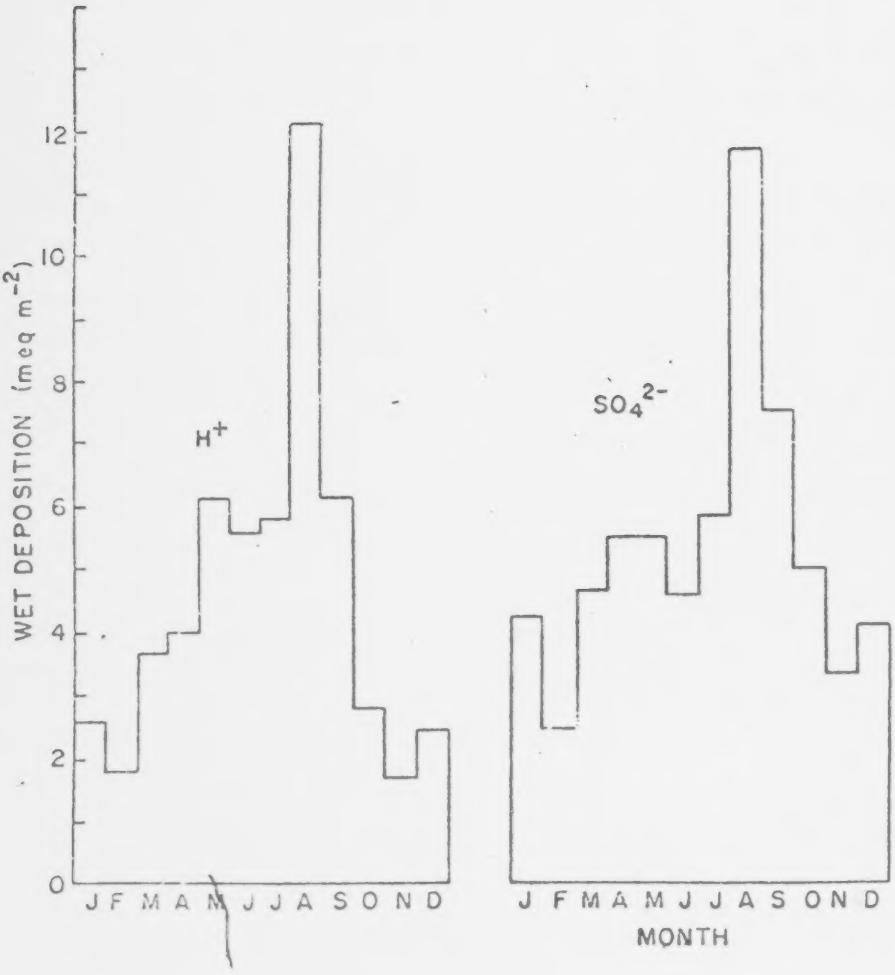

Fig. 3. Monthly wet ion deposition rates at Brookhaven, NY. From (15).

well as the blanketing haze could be averted.

The pending legislations and proposals are primarily aimed toward SO_2 emission control. However, it is also important to consider possible NO_x emission control. Since NO_x emission control technology may in fact be cheaper and simpler than SO_2 control, any effective acid rain control strategy should include NO_x vs. SO_2 emission control trade-offs.

Controlling Emissions

Massive emission reductions such as required under the Mitchell and Moynihan bills are presumed to be accomplished by installation of emission control technology or by substitution of low-emission fuel. The following methods would also be authorized under the pending legislation: least emission dispatch, early retirement of sources, energy conservation, interstate trading of emission reductions, and precombustion cleaning of fuels (16). We shall briefly discuss some of these alternatives, their potentials, prospects, and costs.

Flue gas desulfurization (FGD). There is a voluminous body of literature but meager commercial experience with FGD devices (17,18). In 1979, only 7 percent of the U.S. electric generating plants were fitted with FGD (17).

The basic operative mechanism of FGD is to bring all the post-combustion flue gas in contact with an aqueous solution or slurry of finely ground limestone or lime, with occasional additives of other alkaline substances. This is the so-called wet scrubber method. The alkaline solution/slurry reacts with SO_2 to form a sludge of calcium

sulfite and sulfate (gypsum) which accumulates in the bottom of the reactor. Disposal of the sludge causes considerable engineering, land-use and environmental problems. There are recovery processes available that can regenerate the original sorbent from the spent waste, but their costs are so high that wet scrubbers producing a throwaway sludge are preferred.

Dry scrubbers have been demonstrated but have not yet been used on a commercial scale. Dry scrubbers may cost less and be more suitable in water-deficient areas. Their SO_2 removal yield may be lower than that of wet scrubbers.

Low sulfur coal substitution. Lower sulfur emission rates could be achieved by using low sulfur coal (LSC). Eastern high sulfur coal (HSC) in the average contains 2.7 percent sulfur, western coal 0.7 percent. Limited LSC is also available from eastern coal mines but is used primarily for metallurgic purposes. Western LSC has a 15-25 percent lower calorific content than eastern and midwestern coal, so larger quantities of LSC need be consumed to produce the same amount of heat (or electricity). About 26 Mt of 2.7 percent S coal need to be replaced by 31 Mt of 0.7 percent S coal to achieve a 1 Mt SO_2 reduction.

In general the price of western LSC delivered to an eastern plant is 50 percent greater than eastern HSC. The price differential is even greater for a mine-mouth plant or one holding a long term (low price) contract for HSC. In addition to the price differential, other major barriers to the replacement of HSC by LSC are (a) capital requirements for developing new LSC mines, (b) lack of adequate transport facilities, (c) plant modifications needed to burn lower heat coal, (d) breach of long term contracts, and last but not least (e) the socio-economic

disruption of eastern coal mine districts.

Physical coal cleaning (PCC). There are two basic types of coal cleaning, chemical and physical. Chemical cleaning uses a solvent for extraction of sulfur and could be successful in removing 40 to 60 percent (up to 80 percent in some advanced solvent refined processes) of sulfur from the coal. Chemical cleaning processes are neither commercially available nor economically competitive with other types of sulfur removal. Thus, when coal cleaning is mentioned as an acid rain control method, usually physical coal cleaning is implied.

Physical coal cleaning consists mainly of coal washing by water. Coal washing can remove between 15 and 35 percent of the coal's sulfur. Pulverized coal can be further cleaned using a froth flotation process that takes advantage of the hydrophobic properties of coal and the hydrophilic properties of the impurities. In fact, between 20 to 75 percent (in the Ohio River Basin between 50 to 100 percent) of the coal used by electric utilities is already cleaned, because it reduces the transportation cost, reduces the amount of ash, decreases boiler and flue duct fouling, and reduces the cost of pulverizing coal at the plant. Some studies indicate that over 1 Mty^{-1} SO_2 additionally could be eliminated by PCC (19).

Advanced technology. There are several methods under development that may reduce both SO_2 and NO_x emissions more effectively and at a lower cost than available technology. One of the most promising and least costly methods may be the lime injected multistage burner (LIMB). In this method, lime (or limestone) is injected directly into the combustion chamber where it may absorb up to 40-50 percent of the SO_2. The multistage (low temperature) burner also may significantly reduce

NO_x emissions. The cost of removing one ton of SO_2 by LIMB may be considerably less than by FGD. A related method is the fluidized bed combustor (FBC) where the crushed coal is burned in a bed of crushed limestone suspended by air blown in from below. The limestone "picks up" the sulfur, and the relatively low combustion temperature keeps down the NO_x emissions.

Probably the main reason that LIMB and FBC technology is less advanced than FGD is the perception in industrial circles that these methods will not meet regulatory requirements (e.g., NSPS). A comprehensive and cost-effective acid rain control strategy should foster the development of advanced emission control technology. For even if the SO_2 removal efficiency, say of LIMB, at an individual source is smaller than of FGD, the aggregate emission reductions and cost savings that can be achieved by widespread application of these techniques may be far greater.

Other techniques, still in the early stages of development, have the promise of reducing SO_2 and NO_x emissions while improving the overall efficiency of electric power generation. Among these are coal gasifier combined cycle and magnetohydrodynamics, but it is doubtful that these techniques could contribute significantly to an acid rain control program within the next two decades.

Least emission dispatch (LED). When decreased acid deposition is desired, e.g., during high pollution episodes, the electric generating plants with highest emissions could curtail output or shut down completely while those with lesser emissions (e.g., oil- and gas-fired power plants) take over the excess load. LED is exactly the converse of the generally practiced least cost dispatch, for usually the highest

emission plants (i.e., those using high sulfur coal) generate cheaper electricity. However, LED may in the aggregate be cheaper than installing emission control devices on HSC power plants. In a way, LED is similar to the emission redistribution scheme described above: higher emissions could be permitted in areas and periods that do not contribute significantly to acid deposition in the sensitive areas. The emission redistribution could be accomplished by integrating several utility systems over a regional network, and reducing the load on those units that contribute most severely to acid deposition.

NO_x emission control. Pending legislation is primarily concerned with reducing SO_2 emissions. Since on the average 1/3 of the deposited acid is nitric acid (in some seasons, e.g., winter, and localities, e.g., near metropolitan/industrial areas, the proportion of nitric acid can be even larger), the control of NO_x emissions should not be neglected. In fact, controlling NO_x may be easier and less costly than controlling SO_2. For most of the sulfur is chemically bound to the fuel, while NO_x is primarily formed in the high temperature combustion process by recombining the nitrogen and oxygen of the air. Combustion modifications, such as adjusting the fuel to air ratios, and staged combustion (e.g., the stratified charge internal combustion engine) have shown to be able to reduce NO_x emissions by 40-60 percent (18), and most importantly, could achieve such emission reductions without major modifications or replacement of existing facilities.

Conservation. A surefire acid rain control measure is conservation. Every kWh saved, besides bringing economic advantages, puts 10-20 g less SO_2 and 5-10 g less NO_x through the smokestack of an eastern coal-fired power plant.

Cost assessment. For a reasonably accurate cost figure of any acid rain control strategy one must perform virtually a case-by-case analysis of every major emission source. Some plants are retrofittable with FGD devices, other cannot be so modified for lack of space or disposal areas. Some plants are accessible to LSC, others may be located at a HSC mine mouth. Also, for a reasonable cost assessment, the proper mix of control strategies, such as emission redistribution, least emission dispatch, seasonal or episodic emission control, SO_2 vs. NO_x emission control, etc. need be considered.

To obtain a very approximate estimate of the costs, let us assume that the Mitchell bill goal of reducing SO_2 emissions by 10 Mty^{-1} in 31 states within 10 years would be accomplished with the following mix: 45 percent FGD, 45 percent LSC substitution, and 10 percent PCC.

A survey (17) of 26 operational FGD systems in U.S. coal-fired power plants with average capacity of 360 MW revealed that the mean capital cost (in 1983 dollars) for new installations is $150/kW with a standard deviation (s.d.) of $30/kW. Capital costs include cost of equipment, installation, site development, and start-up. For retrofit, the mean cost is $180/kW, s.d. $60/kW. The annual operating cost for new installations is 10.2 mills/kWh; for retrofit 11.4 mills/kWh (s.d. 3 mills/kWh). Annual costs include cost of raw materials, utilities, operating labor, depreciation, replacement of parts, and interest on borrowed capital.

Let us assume the following parameters: sulfur content in coal 2.7 percent, heat content 12,000 Btu/lb, plant thermal efficiency 0.35, FGD SO_2 removal efficiency 0.8. Thus 1 $kW_e h$ removes 15.4 g SO_2. At 11.4 mills per $kW_e h$, it would cost annually $3.3 billion to remove 4.5

Mty^{-1}.

The cost of substituting LSC for HSC varies greatly from plant to plant depending on the cost of transport and the current price the plant pays for HSC. A very rough price estimate of HSC delivered at an eastern plant is $30 per ton; LSC is $45 per ton. To remove 4.5 Mty^{-1} SO_2, 117 Mty^{-1} HSC (2.7 percent S) need be replaced by 139.5 Mty^{-1} LSC (0.7 percent S). The price differential is about $2.8 billion per year not counting possible capital requirement of rail and storage facilities.

Physical coal cleaning costs vary substantially depending on sulfur content and other characteristics of the coal. Costs per ton SO_2 effectively removed (after combustion) range from $310 to 430 for eastern Midwest coals, $430 to 720 for northern Appalachian coals, and over $1000 per ton for Alabama and southern Appalachian coals (19). Assuming an average price of removing 1 ton SO_2 by PCC of $400, the cost of removing 1 Mty^{-1} is $0.4 billion per year.

Thus, the overall cost of the Mitchell bill with this SO_2 emission reduction scenario would be about $6.5 billion per year with a probable bracket of $5-8 billion, not including the required capital cost. Friedman (14) estimated the cost of the Mitchell bill at $3.3 to $4.1 billion per year. Friedman based his figures on the so-called AIRTEST model developed for the U.S. EPA. No attempt is made here to unravel the sources of the discrepancy between Friedman's estimate and the present analysis. Cost escalations and assumptions about the control technology mix may be the largest factors.

Given the substantial annual cost of proposed acid rain control programs, alternative strategies which emphasize the importance of reducing emissions near sensitive areas, intermittent control and other

techniques hold the promise of significant cost savings compared to the across-the-board continuous reductions exemplified by the Mitchell and Moynihan bills. Quantification of these costs will require a more detailed analysis than has been given here.

Receptor Mitigating Strategies

Lake liming. Liming of aquatic ecosystems has been a relatively longstanding practice for improving fish reproduction in slightly acidic lakes. Large scale programs for the liming of aquatic systems have been under way for some time in Sweden, and are now also being conducted in Norway, Canada, and New York state. The evidence to date suggests that the pH of lakes and streams can be raised to levels that will support fish. The long term effects are not yet established. The lime or limestone mobilizes certain ions, such as aluminum ions, which can be detrimental in certain aquatic systems that have not previously been subjected to carbonate materials. In other aquatic systems there are substantial disruptions of the benthic communities. In addition, the release of the basic material is also difficult to predict, as lime clumps used to promote long-term releases occasionally become coated and ineffective.

The costs of liming can be inferred from the existing programs. By 1986 the Swedish liming program will encompass 200,000 acres of lakes at a cost of $40 million per year, or about $200 per acre of lake per year. Ontario's Ministry of Environment has successfully limed four lakes for about $50 per acre. In the Adirondack Mountains the cost of liming and restocking lakes with bass or trout is estimated to be in the range of

$30 to $400 per acre of lake per year, depending primarily on the accessibility of the lake.

Liming of lakes subject to acidification may be a necessary temporary measure to prevent irreversible damage while acidic deposition is being reduced through emission control programs. As a permanent program for mitigating acid rain effects, however, it has many uncertainties and drawbacks. From a purely chemical view, it would appear more efficient to neutralize the sulfur with lime at the source, rather than at the receptor.

Soil liming. The use of lime to neutralize excess acidity in agricultural soils is a long established and successful practice. However, agricultural crops are not as much endangered by acid rain as forests. Some experiments in Sweden showed that the application of lime to forest soils substantially neutralizes the soils, with one application lasting decades or even centuries. Whether or not this application can reduce the suspected stunting of forest growth is not known, since these experiments have always been conducted in areas where substantial concentrations of gaseous pollutants were also present. Also, it has not been determined from these experiments whether lime interrupts the acidic damage to fine roots or the mobilization of the minerals and metals.

Care must be taken in the application of lime so that the coniferous species, which require relatively acidic soils, will not be placed in too basic conditions, and that the nitrates, which are essential fertilizers, are not drawn away from these terrestrial ecosystems. It is known that relatively little of the basic material makes its way into the surrounding aquatic systems, thus this would not be a cost-effective method of neutralizing lakes. Costs of liming forests are not known, but would be somewhat more expensive than the costs of liming aquatic areas.

Conclusions

Acid rain is difficult and expensive to control. Since vast quantities of both precursors of acid rain--SO_2 and NO_x--are emitted from a multitude of industrial, urban, and transportation sources, restoring the rain acidity to what is postulated to be normal or acceptable is probably not possible without major modifications to all existing emission sources. Modest reduction of the acid deposition rate in certain limited environmentally sensitive areas can be achieved most efficiently by partially curtailing emissions in regions and periods that can be shown to contribute the bulk of acidity to the sensitive areas. This curtailment in the source regions can be accomplished by (a) installing emission control devices on selected emission sources; (b) substituting low sulfur fuel for high sulfur fuel (however, this does not affect NO_x emissions); and (c) importing electricity. All these can only be considered partial remedies; ultimately acid rain will be more effectively controlled by retiring the existing high emission sources and replacing them with new plants that will be equipped with advanced low emission combustion units.

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TARGETED STRATEGIES FOR CONTROL OF ACIDIC DEPOSITION

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SUMMARY

Most of the programs proposed to date to control acidic deposition do not appear to consider the relative costs of achieving emission reductions in different states nor the relative contributions of emissions in different states to acidic deposition at sensitive receptor sites. This may be because control cost models and atmospheric transport models are not presently verifiable to a degree necessary to support confident conclusions about the effects of emission control strategies. However, these models can be usefully employed to discern relative merit among different strategies, at broad levels of source/receptor aggregation.

The purpose of this analysis is to show that, if one acknowledges the considerable uncertainty associated with currently available tools, control strategies can be designed to achieve specific objectives with considerably improved efficiency compared to the strategies under consideration by Congress. In particular, this analysis examines cost-effective strategies to minimize acidic deposition in certain "targeted" regions that are considered vulnerable to acidic deposition.

In an effort to demonstrate the potential economies achievable by applying a targeted approach to control, several versions of the two Senate Bills S.768 and S.769 are evaluated in terms of annual control costs, SO_2 emission reductions, and sulfur deposition rates in nine sensitive regions. The sensitive regions are: Boundary Waters, Algoma, Muskoka, Quebec, Southern Nova Scotia, Vermont/New Hampshire, Adirondacks, Pennsylvania, and the Smoky Mountains.

The results are striking. The control measures proposed in the legislation (assuming intrastate emissions trading) are relatively expensive because of the economically inefficient allocation of emission reductions

among the states. Regional cost optimization over the eastern U.S. (interstate emissions trading) can reduce costs by more than 50%. Regional cost optimization also increases the deposition reduction achieved in critical sensitive areas.

A targeted approach to control is analyzed that would achieve the same reduction in sulfur deposition in one of the most critical sensitive regions, the Adirondacks, as the bills would achieve, at minimum cost. This could lower the cost of achieving that level of deposition reduction in the Adirondacks by between 75 and 90%, with the estimate depending upon which of several atmospheric transport models are used.

The targeted strategy designed to reduce sulfur deposition in the Adirondacks at minimum cost is also effective at reducing deposition in other sensitive regions in the northeastern U.S. and southeastern Canada. However, this approach does not yield as great a deposition reduction in the Boundary Waters, Algoma, and Smoky Mountains regions as do the legislative proposals. This may not be critical in the near future for the first two of these areas, because current deposition rates are relatively low. The Smoky Mountains, on the other hand, might merit a second targeted strategy, because deposition rates are relatively high there.

The targeted strategy is sensitive to meteorological variability. A factor of two difference in control costs resulted from model runs which used meteorology from two different time periods. Meteorological variability in areas close to the sensitive receptor regions is particularly important.

Targeted strategies can also be designed to meet multiple objectives, such as achieving desired reductions in deposition in many sensitive regions at minimum cost. This analysis shows that wet deposition of sulfate can be reduced to levels below the value recommended by a number of scientists, 20 kg

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ha^{-1} yr^{-1}, in all nine of the sensitive regions, at a cost slightly less than the cost of an unoptimized S.769. A target of 30 kg ha^{-1} yr^{-1} can be achieved in all sensitive regions with approximately an 80% reduction in control costs compared to S.769 and the 20 kg targeted option.

The control costs evaluated in this analysis assume that fuel switching is an allowable option for electric utilities to achieve emission reductions, in addition to technological control methods (use of flue gas desulfurization) and coal washing. Fuel switching, however, has the potential to disrupt established markets of coal supply, causing miner dislocation and secondary cost impacts. To avoid these impacts, control programs have been proposed that would necessitate the use of flue gas desulfurization technology alone to achieve the required emission reductions. This analysis shows that costs would increase by 6-21%, depending upon the control strategy considered, if fuel switching is not a permissible compliance option.

In conclusion, this analysis indicates that, if reducing sulfur deposition in the northeastern U.S. and southeastern Canada is the primary objective of an acid deposition control program, then there is a large potential for cost savings by using a targeted approach. The quantitative findings of the study are summarized in the following table.

Summary Table

Strategy	Annual Cost[a] ($ billion)	Reduction in Annual SO_2 Emissions[b] (million tons)	Reduction in Total Sulfur Deposition[c] in the Adirondacks (%)
S.768			
Intrastate Trading	4.2	9.0	23 (21-26)
Interstate Trading	1.9	9.0	26 (26-31)
Deposition Optimized	0.9 (0.4-1.1)	5.0 (1.9-5.5)	23 (21-26)
Intrastate (FGD only)	4.6	9.0	23
Interstate (FGD only)	2.3	9.0	27
S.769			
Intrastate Trading	12.9	14.9	40 (38-46)
Interstate Trading	6.0	15.0	46 (45-57)
Deposition Optimized	3.0 (1.3-3.2)	9.8 (4.2-10.3)	40 (39-46)
Intrastate (FGD only)	13.6	14.9	40
Interstate (FGD only)	7.0	15.0	47
20 kg Target[d] Loading	11.9	16.2	56
30 kg Target[d] Loading	2.5	9.4	29

Numbers in parentheses represent a range of results for six atmospheric transport models.

[a] In real, levelized 1980 dollars
[b] In English short tons
[c] Modeled using ASTRAP with 1980 meteorological data
[d] Measured in kg ha^{-1} yr^{-1} of wet sulfate

Note: Costs are optimized using the techniques described in the appendices. Real-world costs would be expected to be greater than these theoretical least costs.

INTRODUCTION

The task of properly designing and evaluating acidic deposition control strategies requires the ability to:

1. Define dose-response relationships relating different deposition rates of sulfur and nitrogen compounds to different levels of adverse effects;
2. Relate deposition rates of sulfur and nitrogen compounds to emission rates, for sources close to and hundreds of miles from deposition sites; and
3. Quantify the economic values of different levels of adverse effects and the costs of a range of emission control alternatives for sulfur and nitrogen compounds.

With these capabilities in hand one would be able to calculate the degree of emission control needed to achieve a set of desirable results and to determine the net benefit to society of any particular strategy. The Interagency Task Force on Acid Precipitation is currently conducting research directed at these needs. However, these essential analytical capabilities are not yet developed to the desired degree.

In order to address the acidic deposition issue, fourteen bills were introduced in the 97th Congress, and so far (to October 1, 1983) eleven bills have been introduced in the 98th Congress. The majority of these bills call for reductions in annual sulfur dioxide emissions ranging between 6 and 14 million tons. The legislators proposing these bills obviously believed that the lack of quantitative cause-and-effect relationships was insufficient reason to delay action.

The bills require states to reduce their sulfur dioxide emissions according to various allocation schemes, in order to achieve the mandated

total reductions. Various options for financing the control programs have been proposed. However, the actions proposed by the bills do not appear to be based on analysis according to the three requirements stated at the beginning of this paper. The driving mechanism in each case is to require reductions at the state level in approximate proportion to current state-level emissions in the eastern U.S. Only limited consideration is given to the relative costs of achieving emission reductions in different states and none is given to the relative contributions of emissions in different states to acidic deposition at sensitive receptor sites.

The purpose of this paper is not to advocate any emission control program, but rather to close the gap between the ideal evaluation method, which is currently impossible to define, and the very simple rules of thumb followed by the bills. The analysis shows that, if one accepts the considerable uncertainty associated with currently available tools, control strategies can be designed to achieve specific objectives with considerably improved efficiency compared to the strategies under consideration by Congress. In particular, this analysis examines cost-effective strategies to minimize acidic deposition in certain "targeted" regions that are considered particularly vulnerable to this form of environmental pollution.

This concept is not new. Similar control strategies have been recommended in the literature. They recognize that emissions in some states can be controlled at much lower cost than in other states and that contributions to deposition in receptor regions from emitting sources or states are inversely related to distance of separation. Trisko[1] has recommended "a phased program of source controls, starting with nearby ... sources, and expanding later to more distant ... sources as necessary ..." Senator Richard G. Lugar of Indiana has stated[2] that "although Midwestern

emissions are undeniably greater than those in the Northeast, reductions in the Midwest may have a lesser effect because they are 500 to 900 miles from the affected areas. Thus, the cost of the cleanup can be lowered by targeting reductions as close as possible to the affected areas of the Northeast. The Midwest would gain through reduced costs for cleanup, the Northeast by achieving maximum benefits for its sensitive ecosystems." An earlier analysis by Fay et al.[3] suggested that "considerably lower costs could be realized in an emission control scheme that is the more stringent the nearer the sources are to the environmentally sensitive areas."

Improved efficiency (on cost or deposition grounds) inevitably leads to reduced equity (that is, several states bear a much larger share of the burden of emission reductions than the remaining states). This fact was recognized by the legislators who drafted bills calling for uniform emission control requirements for all states. However, if cost (who pays) can be decoupled from emission reduction (where controls are installed) then this problem is largely obviated. Several proposals by Senator Danforth,[4] Senator Glenn,[5] and Representatives Sikorski, Waxman, and Gregg[6] attempt to achieve this decoupling by partially financing the emission control program through taxes on electricity generation, loan guarantees, or tax credits, distributed among all states in the region or the nation.

Another major problem concerns the confidence that can be placed in our ability to determine the relative contributions of emissions in different states to acidic deposition in receptor regions. The executive summary of the recent report on acid deposition by the National Research Council acknowledges that "... as a rule of thumb ... the farther a source is from a given receptor site, the smaller its influence on that site will be per unit mass emitted."[7] However, the executive summary advises "caution in using deterministic

[long-range transport] models to project changes in patterns of deposition on the basis of changes in patterns of emissions of precursor gases."[8] This is clarified somewhat in the body of the report by the statement that ... "current models have not provided results that give confidence in their ability to translate SO_2 emissions from specific sources or localized groupings of sources to specific sensitive receptors"[9] (emphasis added). The issue can perhaps be summarized by stating that uncertainty in the simulations with current long-range transport models of contributions of single or local groups of sources to deposition at specific receptor sites cannot yet be rigorously determined, and that contributions from sources aggregated to a higher level (state or region) should also be viewed with caution. This analysis looks at emission changes at the state level and does not attempt to assign culpability to individual sources of emission. The results of this analysis are so striking that it would appear to be unwise to design emission control strategies without the use of these atmospheric transport models, notwithstanding the level of uncertainty associated with them.

This article presents a quantitative analysis of targeted emission control strategies that comes as close to simulating the ideal evaluation scheme discussed at the beginning of the article as currently available tools and the current state of scientific knowledge will permit. The limitation on available tools means that a number of potentially useful strategies (such as controlling NO_x emissions) cannot be satisfactorily evaluated. Additionally, because of quite large uncertainties in these tools, substantial confidence cannot be placed in their absolute numerical outputs. However, they should be useful to discern relative merit among certain strategies, particularly strategies having fundamentally different regulatory approaches.

The analysis presented here makes use of four key elements:

1. A program to calculate emission reduction requirements for each state for a given reduction formula provided by a Congressional bill or similar policy.
2. A model to rank order, by cost, various opportunities to reduce sulfur dioxide emissions in each state, for all emitting source sectors.
3. A set of transfer matrices, derived from current long-range atmospheric transport models, relating 40 emission source areas to 9 sensitive receptor areas, to provide a relative measure of source/receptor relationships.
4. The locations of sensitive receptor areas, as indicators of the geographical targets for deposition reduction and certain target loadings for receptors, as measures of the desirable magnitude of deposition reductions.

These elements are described in detail in Appendices A-D, respectively. What follows is a presentation of the results obtained by using these methods to evaluate certain targeted control strategies. It must be emphasized that the optimized costs presented are theoretical least costs. Actual costs incurred in implementing any of the strategies can be expected to be higher.

RESULTS

1. The Senate Bills

Two recently proposed control programs form the starting point for this analysis: the Senate Bills S.768 and S.769. These bills were both introduced into the 98th Congress on March 10, 1983, by Senator Robert T. Stafford. The bill S.768, formerly the bill S.3041 reported out of the Senate Committee on Environment and Public Works during the 97th Congress, calls for a reduction of 8 million tons in annual sulfur dioxide emissions in the eastern U.S. The bill S.769 calls for a reduction of 12 million tons in such emissions. All emissions presented in this paper are measured in English short tons, unless otherwise stated.

With growth offsets, these reductions become 9.0 and 14.9 million tons per year, respectively. The reductions are large (41% and 68%, respectively), compared to 1980 annual SO_2 emissions in the East of 21.8 million tons.[10] The first step in this analysis was to allocate total emission reductions to the states using the formulas prescribed in the bills. Appendix A discusses this in detail and presents state-level information on the requirements of the bills.

Assuming cost optimization within each state (so-called "intrastate trading"), the costs of these bills are calculated to be $4.2 and $12.9 billion per year, respectively. All costs presented in this article are in real, levelized 1980 dollars, unless otherwise stated. Appendix B discusses the methodology used in the cost calculations. ICF, Inc.,[11] has estimated the cost of S.768 to be $3.1 billion per year; and the Energy Information Administration[12] has estimated its cost to be $3.6 billion per year -- both in levelized 1980 dollars. Table 1 shows the costs of these bills.

2. Cost-Optimized Bills

The bills are indifferent among states as to how costly it is to achieve SO_2 emission reductions. They are also indifferent, geographically, as to which states are required to reduce emissions. Therefore, the question arises as to whether the total emission reduction could be achieved at lower cost by optimizing over the entire region -- that is, by achieving the reductions in those states where it is most cost-effective to apply additional controls. This is equivalent to an "interstate trading" option.

Regional cost optimization of the bills S.768 and S.769 yields costs of $1.9 billion and $6.0 billion per year, respectively. These are reductions of 55% and 54%, respectively, compared with the unoptimized bills. Thus, control costs could be reduced by about half, if the bills were regionally optimized on the basis of least cost. These costs are also shown in Table 1.

Of course, the regional distribution of emission reductions among the states now changes (see Appendix A), possibly necessitating a scheme for more equitable distribution of costs (see Introduction).

3. Baseline Deposition

In order to be able to evaluate targeted control strategies, one needs to be able to calculate changes in deposition resulting from a control measure, relative to a baseline corresponding to deposition in the absence of control measures. Source/receptor transfer matrices from six models of atmospheric transport and deposition, exercised by Work Group 2 in research related to the Memorandum of Intent (MOI) on Transboundary Air Pollution between the United States and Canada (see Appendix C), were used to generate baseline deposition data for the years 1980 and 2000, using emissions projections also generated for the U.S.-Canadian MOI study.[10] Table 2

summarizes the baseline deposition data evaluated for each of the six models for nine sensitive receptor regions (see Appendix D). The extent of agreement among the models can be judged from this table. Deposition rates for 1980 and 2000 are not greatly different for a given model, because SO_2 emissions are calculated as being almost equal for 1980 and 2000. In the results that follow, changes in deposition resulting from a control scenario are generally expressed as percentage changes in deposition in the year 2000 over the range of models, in order to minimize the differences among the models.

4. Deposition Reductions Achieved by the Bills

Using the six transfer matrices, the emission reductions achieved by the bills at the state level can be translated into changes in total sulfur deposition in the nine sensitive regions. It should be recognized that these sensitive regions are meant to be representative and not a complete listing of all sensitive regions. Table 1 shows percentage reductions in deposition achieved in each region by S.768 and S.769, for both intrastate and interstate trading options. The ranges presented bound the results obtained using the six models. The ranges are not intended to imply confidence intervals or accuracy estimates, but rather to demonstrate the extent of disagreement among the models. By expressing reductions as a percentage of the baseline projection, a significant fraction of the model disagreement is removed. In fact, the models agree better than the width of the range suggests; in many cases there is a single, outlying estimate (usually from the ENAMAP model).

Table 1 shows that total sulfur deposition in sensitive regions in the Northeast (Regions 4-7) is reduced by amounts in the range of 7-26% under S.768 and 13-46% under S.769. Deposition reduction in the Pennsylvania and Smoky Mountains regions is somewhat higher (34-40% and 58-65%, respectively).

A significant finding is that the cost-optimized bills, in addition to reducing control costs by about half, actually achieve greater deposition reduction in some of the critical sensitive areas than do the bills: 9-31% in the Northeast under a cost-optimized S.768 and 17-57% under a cost-optimized S.769. Deposition reduction in Pennsylvania and the Smoky Mountains is unchanged. Only the Boundary Waters and Algoma areas fare marginally worse, but here the current deposition rate is relatively low.

5. Deposition-Optimized Bills

The six transfer matrices were then used to optimize the emission reductions in order to achieve desired deposition reductions in sensitive receptor regions at minimum cost. A single "target" region was used in this case: the Adirondacks. The cost and deposition models were used to determine the least-cost distribution of emission reductions that would achieve the same amount of reduction in total sulfur deposition in the Adirondacks as would the bills.

The results are striking. At a cost of between $0.4 and $1.1 billion per year, the same level of deposition reduction (21-26%) can be achieved in the Adirondacks as would be achieved by S.768 (which costs $4.2 billion per year). Again, the cost ranges correspond to the results obtained using the six different transport models. Costs savings would therefore be between 75 and 90%. The necessary reduction in SO_2 emissions is 1.9-5.5 million tons per year, as compared with 9 million tons per year under the bill.

Similarly, at a cost of $1.3-3.2 billion per year, the same deposition reduction (39-46%) can be achieved in the Adirondacks as S.769 (which costs $12.9 billion per year) would achieve. Cost savings in this case are again between 75 and 90%. The necessary reduction in SO_2 emissions is

correspondingly lower than required by the bill (4.2-10.3 million tons per year vs. 14.9 million tons per year). Also shown in Table 1 are the deposition-optimized strategies that achieve the same levels of deposition reduction as the cost-optimized bills.

It is generally found to be true that the deposition-optimized strategies perform well in the other sensitive regions in the Northeast (4, 5, 6, and 7) when optimized for deposition reduction in the Adirondacks. However, significantly lower levels of deposition reduction are achieved in the Boundary Waters, Algoma, and Smoky Mountains regions under this targeted strategy. Because current loadings are relatively light in the first two areas, only the Smoky Mountains region appears to be of immediate concern in this regard. The extent to which this concern is of major importance can perhaps be related to the relative sensitivities of these regions (that is, the Adirondacks compared to the Smoky Mountains) and the trade-off with cost savings. Multiple-targeted strategies related to target loadings for all of these regions are evaluated later in this article.

Figure 1 brings the targeted strategy data into sharp focus for the Adirondacks. It clearly illustrates the potential for a targeted approach to provide significant cost advantages if the objective is to reduce sulfur deposition in the Adirondacks (and, with little loss of effectiveness, the northeastern U.S., and southeastern Canada). As an illustration, the targeted strategy is clearly least expensive to achieve, say, a 40% reduction in deposition in the Adirondacks. In contrast, an interstate trading strategy costs about 50% more and an intrastate trading approach costs about 4 times as much. These curves were constructed from results obtained using the transfer matrix for the ASTRAP model with 1980 meteorological data.

Shifts in state-level emission reduction patterns in going from the bills with intrastate trading to interstate trading and deposition optimized approaches are presented in Appendix A. Figures 2 and 3 illustrate how the reduction requirements shift to states with relatively inexpensive cleanup opportunities (under interstate trading) and to states with a combination of relatively inexpensive cleanup opportunities and upwind proximity to the Adirondacks (in deposition-optimized strategies). For these figures, the ASTRAP model with 1980 meteorology was chosen (see discussion later in the paper).

6. Meteorological Variability

The ASTRAP matrices that were used in the U.S.-Canadian MOI study were based on two months of meteorological data: January and July of 1978. It was known that July 1978 experienced reasonably normal climatological conditions, but that January 1978 was very wet in the northeastern U.S. The possibility was recognized, therefore, that the wet deposition contribution from source states in the Northeast to receptor regions in the Northeast during the latter month might have been unusually high. It is important that meteorological data used in these analyses be as "typical" as possible, for projections are being made of the future effects of control programs where reliance must be placed on historical meteorology. In particular, it was believed that targeted strategies might be susceptible to meteorological variability.

Tables 3 and 4 show the projected deposition reductions for the two bills using the ASTRAP model with three sets of meteorological data: the two months of 1978 data used in the MOI study, and 12 months of data for each of 1980 and 1981.

The patterns of reductions in total sulfur deposition resulting from the bills show no large differences. The deposition reduction is slightly greater, in percentage terms, in the Boundary Waters area and lower in Algoma, when 1980 and 1981 data are used. The two more recent data sets produce very similar patterns of deposition.

However, there is a significant difference between the 1978 and 1980/1981 data sets when deposition-optimized strategies are devised. The first observation is that cost estimates approximately double when the more recent data are used, for either S.768 or S.769. The required emission reductions increase by about 130% for S.768 and by about 60% for S.769. However, costs and emission reductions are still considerably less than in the cases of the unoptimized bills. The increased emission reductions with 1980/1981 data also cause deposition to be correspondingly reduced to a greater extent in the other sensitive regions, particularly those regions away from the Northeast (i.e., 1, 2, 3, 8, and 9).

The reason for these differences becomes clear if the state contributions to deposition reductions in the Adirondacks under optimized conditions are compared across the three meteorological data sets (Table 5). In the 1978 case, New York State itself would contribute over 60% of the deposition reduction necessary in the Adirondacks under a deposition-optimized S.768. Using 1980 or 1981 data, the deposition-reduction contribution from New York State is no greater than 30%. This arises because the unit matrix deposition value for New York State -> Adirondacks for 1978 is about 2-1/2 times what it is for 1980 or 1981 (also shown in Table 5). As suspected, this corresponds to an unusually wet January of 1978, which greatly increases the rate of wet deposition in New York State, especially when only two months of data are used. Because New York State contributes less in 1980 and 1981,

greater contributions have to be "raised" from more distant states like Ohio, West Virginia, and Missouri, thereby increasing the necessary emission reduction and increasing the cost.

It can be concluded from this comparison that targeted control strategies are susceptible to meteorological variability, particularly in the vicinity of the target regions. However, meteorological variability, even in extreme cases, does not detract from the cost advantage of a targeted strategy relative to an unoptimized strategy. It does point out the need for long-term averaged (over at least several years) meteorological data to improve the predictive capabilities of the models in this kind of application. For the purposes of this analysis, we place greater confidence in the ASTRAP results using 1980 or 1981 data than using the MOI 1978 data. The 1980 data set is chosen for several of the sensitivity analyses in this paper, to be consistent with the cost modeling and the current baseline deposition data. As a closing observation, note that the range of costs and emission reductions obtained using one model (ASTRAP) with different meteorology still falls within the range of results obtained using several different models with the same meteorology.

7. Target Loading Strategies

As part of work conducted pursuant to the U.S.-Canadian MOI, several scientists proposed a wet deposition rate of 20 kg $SO_4^=$ Ha^{-1} yr^{-1} as desirable to protect all but the most sensitive lakes. "Most sensitive" was postulated to be those lakes with alkalinity less than 40 microequivalents/liter. Other scientists, on the other hand, concluded that no target loading could be derived for lakes in general, and that for the specific areas where data were available, 30 kg $SO_4^=$ Ha^{-1} yr^{-1} was adequate. Various other target loadings

have been proposed (see Appendix D), but none have any wide acceptability at this time. For the purposes of demonstrating the applicability of the method, and also to give some indication of the implications of these levels of target loadings, two strategies were examined which would keep wet deposition in all of the sensitive regions below 20 (30) kg $SO_4^=$ Ha^{-1} yr^{-1}.

Immediately, the scientific footing becomes less sure. Whereas in the previous analyses we have not had to venture beyond relative changes in deposition between a baseline scenario and a control scenario, we now have to place confidence in the ability of the deposition models to predict absolute levels of deposition. In preliminary comparisons with measurements of annual wet deposition of sulfur in four U.S. and Canadian monitoring networks, ASTRAP simulations explained almost 75% of the observed variance in 1980, which is encouraging. Nevertheless, the results presented in this section should be viewed with greater caution than the results in the earlier sections.

The baseline deposition data for the three ASTRAP data sets are shown in Table 6. These enable a set of target reductions to be established (in kg total S Ha^{-1} yr^{-1}) which would bring the year 2000 wet deposition values in each sensitive region below 20 or 30 kg $SO_4^=$ Ha^{-1} yr^{-1}. Table 7 shows these targets evaluated using the ASTRAP model with 1980 meteorology. The primary need for deposition reductions is in Pennsylvania and the Smoky Mountains. The 20 kg target also requires significant reductions in the Adirondacks, Muskoka, and Vermont New Hampshire. In other cases, deposition is either projected to be lower than the targets without any additional controls being applied; or it is expected that the targets will be readily met by reductions necessary to achieve targets in other regions.

Multiple-targeted, least-cost strategies were then calculated which would, at minimum, achieve each of these deposition targets in each sensitive

region. The cost to keep wet deposition below 20 kg $SO_4^=$ Ha^{-1} yr^{-1} was estimated to be $11.9 billion per year, corresponding to a 16.2 million tons reduction in annual SO_2 emissions. The cost to keep wet deposition below 30 kg $SO_4^=$ Ha^{-1} yr^{-1} was estimated to be $2.5 billion per year, corresponding to a 9.4 million tons reduction in annual emissions of SO_2. Table 7 compares these strategies with the bills assuming intrastate and interstate trading.

The results show that, with targeted optimization, all sensitive regions could receive the more stringent level of protection (20 kg $SO_4^=$ Ha^{-1} yr^{-1}) at a cost slightly less than the estimated cost of S.769 as currently constituted. The cost is relatively high because of the target imposed in the Muskoka region (a reduction of 12 kg S Ha^{-1} yr^{-1}). To achieve this reduction from U.S. sources alone is very expensive; a reduction in emissions from Canadian sources would likely be more cost effective. A lower level of protection in the sensitive regions (30 kg $SO_4^=$ Ha^{-1} yr^{-1}) could be achieved at about 19% of the cost of meeting S.769 and at about 21% of the cost of meeting a 20 kg target.

8. No Fuel Switching Permitted

One of the potential drawbacks to acid deposition control strategies that mandate a certain level of total emission reductions without specifying the technological means of achieving those reductions is that fuel switching becomes an attractive option for electric utilities. Modest levels of reduction can be obtained at relatively low cost by switching to fuels of lower sulfur content, rather than installing costly flue gas desulfurization systems. This leads to disruptions in the patterns of fuel supply. In particular, the midwestern high-sulfur coal industry may be adversely

affected. Political opposition to such strategies is therefore engendered among certain states and certain interest groups.

To counteract this negative impact, strategies have been proposed which imply or require the use of flue gas desulfurization alone to achieve the required emission reductions. Costs of the strategies are correspondingly higher. In order to investigate the cost and deposition implications of a "mandatory FGD" scenario, a parallel set of state-level emission reduction opportunities was compiled, but without the option of fuel switching for electric utilities. Industrial fuel switching was still permitted. Appendix B discusses this option in more detail. Table 8 shows the results obtained for S.768 and S.769 when fuel switching is not permitted.

As expected, costs increase, but not as much as might perhaps have been expected (6-21% of base costs). Three observations are appropriate. First, the lower the reduction requirement, the greater the cost difference in percentage terms (9% for S.768 vs. 6% for S.769). This is because fuel switching becomes increasingly less able to meet the emission reduction requirements as the latter rise; FGD becomes the only feasible option at high levels of reduction. Second, the cost difference is greater under cost-optimized strategies than under the unoptimized bills (21% vs. 9% for S.768; 17% vs. 6% for S.769). This is because <u>all</u> opportunities for cheaper fuel switching can be utilized before the need for FGD, when constraints at the state level are lifted. Third, the patterns of deposition reduction do not significantly change when the mandatory FGD condition is imposed on the cost-optimized strategies. This suggests little variation among the states in the <u>relative</u> cost differences between fuel switching and FGD.

Figures 4 and 5 show the least-cost regional control curves with and without utility fuel switching. Costs and emission reductions for interstate

trading options are points on these curves. Figure 4 shows the complete curve for the ARMS region up to a theoretical maximum of 21.8 x 10^6 English short tons per year removed. These curves are compiled by merging the state data as contained in Tables B-6 and B-7 and plotting cumulative emission reductions versus cumulative costs. The curves are made up of individual points associated with the many state/source category combinations.

In Figure 4, one can see the "knee" in the curves, at about 16.5 x 10^6 tons/year, where the "reasonably reducible" SO_2 becomes exhausted and the "$4400/ton" SO_2 becomes utilized (see Appendix B). Figure 5 shows an expansion of the curves in Fig. 4 in the region of 0-8 million tons removal.

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Table 1 Summary of the Bills and Their Optimized Alternatives

	Annual Cost ($ billion)	Emission Reduction (MM tpy)	Range of Deposition Reductions (% of 2000 Deposition)								
			DR1	DR2	DR3	DR4	DR5	DR6	DR7	DR8	DR9
S.768											
A	4.2	9.0[a]	19-31	24-39	13-31	7-23	8-23	17-23	21-26[b]	34-36	36-40
B	1.9	9.0[a]	12-16	21-31	13-31	9-25	16-28	20-28	26-31[c]	39-42	32-37
C	0.4-1.1	1.9-5.5	0-11	0-20	3-20	4-18	9-21	17-22	21-26[b]	19-34	1-11
D	0.6-1.3	2.5-6.1	0-12	1-21	4-21	5-21	12-25	19-26	26-31[c]	26-39	1-13
S.769											
A	12.9	14.9[d]	29-48	35-57	21-49	13-39	21-41	32-41	38-46[e]	58-59	59-65
B	6.0	15.0[d]	26-41	31-46	22-49	17-43	35-51	35-50	45-57[f]	62-65	60-67
C	1.3-3.2	4.2-10.3	0-21	2-35	8-34	8-34	19-38	31-38	39-46[e]	41-57	4-24
D	3.1-4.7	6.0-11.9	0-72	4-40	13-38	12-43	30-45	34-48	45-58[f]	49-65	8-33

a-f Items with the same superscript were held constant for a given optimization. Small numerical differences arise from the computational techniques used.

A = Intrastate Trading
B = Interstate Trading
C = Least-cost Optimization to give same Adirondacks Deposition as A
D = Least-cost Optimization to give same Adirondacks Deposition as B

DR1 = Boundary Waters
DR2 = Algoma
DR3 = Muskoka
DR4 = Quebec
DR5 = S. Nova Scotia
DR6 = Vermont/New Hampshire
DR7 = Adirondacks
DR8 = Pennsylvania
DR9 = Smokies

Table 2 Baseline Deposition Data for Nine Sensitive Regions in the Year 1980 and 2000, Evaluated Using Six Transport Models

	Deposition in Year 1980 (Kg S Ha^{-1} yr^{-1})								
	DR1	DR2	DR3	DR4	DR5	DR6	DR7	DR8	DR9
AES	3.26	13.72	29.53	13.96	10.92	24.10	25.77	60.33	33.72
ASTRAP	3.03	11.50	20.84	18.29	9.89	21.57	28.13	50.95	21.43
MCARLO	2.02	12.50	33.46	23.45	11.89	23.07	32.65	55.93	24.60
ENAMAP	0.99	2.38	22.63	8.80	7.56	12.91	17.91	48.64	19.49
OME	2.04	6.30	10.61	7.72	9.10	12.04	12.91	33.77	15.50
UMACID	0.81	3.97	15.37	7.29	6.09	8.20	12.07	31.41	20.41

	Deposition in Year 2000 (Kg S Ha^{-1} yr^{-1})								
	DR1	DR2	DR3	DR4	DR5	DR6	DR7	DR8	DR9
AES	4.19	14.77	28.19	12.98	10.15	22.38	25.21	62.97	38.60
ASTRAP	3.93	13.48	21.37	16.78	9.64	20.63	27.27	52.92	25.52
MCARLO	2.49	13.85	32.18	22.07	11.11	22.09	32.14	58.28	28.28
ENAMAP	1.19	2.29	19.85	7.26	6.45	11.72	16.91	50.49	21.44
OME	2.43	7.00	10.74	7.23	8.85	11.30	12.75	35.33	17.77
UMACID	0.99	4.50	14.90	6.62	5.71	7.88	11.86	32.67	23.16

DR1 = Boundary Waters
DR2 = Algoma
DR3 = Muskoka
DR4 = Quebec
DR5 = S. Nova Scotia
DR6 = Vermont/New Hampshire
DR7 = Adirondacks
DR8 = Pennsylvania
DR9 = Smokies

Table 3 Summary of S.768 and Its Deposition-Optimized Alternative Using ASTRAP with Different Meteorological Data

	Annual Cost ($ billion)	Emission Reduction (MM tpy)	Reduction in Deposition (% of 2000 Deposition)								
			DR1	DR2	DR3	DR4	DR5	DR6	DR7	DR8	DR9
BILL											
ASTRAP/1978[d]	4.24	8.98	22	39	31	20	23	23	24[a]	34	38
ASTRAP/1980[e]	4.24	8.98	26	33	26	21	20	20	23[b]	35	39
ASTRAP/1981[e]	4.24	8.98	26	34	27	22	21	21	24[c]	34	39
DEPOSITION OPTIMIZED											
ASTRAP/1978[d]	0.44	2.15	1	4	6	10	14	18	25[a]	19	1
ASTRAP/1980[e]	0.92	5.01	10	15	15	18	20	21	23[b]	31	8
ASTRAP/1981[e]	0.92	4.98	9	14	16	19	20	22	24[c]	30	8

[a-c] Items with the same superscript were held constant for a given optimization. Small numerical differences arise from the computational techniques used.

[d] Based on 2 months of meteorological data (January and July)

[e] Based on 12 months of meteorological data

DR1 = Boundary Waters
DR2 = Algoma
DR3 = Muskoka
DR4 = Quebec
DR5 = S. Nova Scotia
DR6 = Vermont/New Hampshire
DR7 = Adirondacks
DR8 = Pennsylvania
DR9 = Smokies

Table 4 Summary of S.769 and Its Deposition-Optimized Alternative Using ASTRAP with Different Meteorological Data

	Annual Cost ($ billion)	Emission Reduction (MM tpy)	Reduction in Deposition (% of 2000 Deposition)								
			DR1	DR2	DR3	DR4	DR5	DR6	DR7	DR8	DR9
BILL											
ASTRAP/1978[d]	12.85	14.94	32	57	49	33	41	41	43[a]	58	63
ASTRAP/1980[e]	12.85	14.94	38	50	41	35	38	35	40[b]	58	63
ASTRAP/1981[e]	12.85	14.94	39	51	43	38	38	37	41[c]	57	63
DEPOSITION OPTIMIZED											
ASTRAP/1978[d]	1.48	6.13	8	20	23	22	29	35	43[a]	41	7
ASTRAP/1980[e]	2.98	9.83	19	33	32	32	37	36	40[b]	54	19
ASTRAP/1981[e]	2.96	9.73	17	33	35	35	37	38	41[c]	53	18

[a-c] Items with the same superscript were held constant for a given optimization. Small numerical differences arise from the computational techniques used.

[d] Based on 2 months of meteorological data (January and July)

[e] Based on 12 months of meteorological data

DR1 = Boundary Waters
DR2 = Algoma
DR3 = Muskoka
DR4 = Quebec
DR5 = S. Nova Scotia
DR6 = Vermont/New Hampshire
DR7 = Adirondacks
DR8 = Pennsylvania
DR9 = Smokies

Table 5 The Six Major Source States Contributing to Adirondacks Deposition in S.768 and Optimized S.768 Using Alternative ASTRAP Data

	Reduction in Deposition (Kg S Ha^{-1} yr^{-1})					
	1978[a]		1980[b]		1981[b]	
	BILL	OPT.	BILL	OPT.	BILL	OPT.
	OH 1.45	NY 4.36	OH 1.45	NY 1.67	OH 1.53	NY 2.00
	PA 1.35	PA 1.52	PA 0.99	PA 1.67	PA 1.13	PA 1.90
	IL 0.57	OH 0.53	WV 0.68	OH 1.15	WV 0.76	OH 1.21
	IN 0.46	IN 0.17	MI 0.41	WV 0.48	IL 0.48	WV 0.53
	WV 0.43	WV 0.07	IL 0.40	MI 0.18	NJ 0.37	MO 0.22
	MI 0.40	IA 0.06	IN 0.31	MO 0.17	MI 0.36	IL 0.18
	.	.	.	.	.	.
	.	.	.	.	.	.
	.	.	.	.	.	.
TOTAL	6.45	6.78	5.92	5.93	6.66	6.74

[a]Based on 2 months of meteorological data (January and July)
[b]Based on 12 months of meteorological data

Note: UNIT MATRIX DEPOSITION VALUES (Kg S Ha^{-1} yr^{-1} $(TgS)^{-1}$) FOR NY STATE ⟶ ADIRONDACKS:

	1978	1980	1981
WET	9.64	2.75	3.42
DRY	9.37	4.54	5.29

Table 6 Baseline Deposition Data for Nine Sensitive Regions in the Years 1980 and 2000 Using Alternative ASTRAP Matrices

	DR1	DR2	DR3	DR4	DR5	DR6	DR7	DR8	DR9
	Deposition in Year 1980 (Kg S Ha^{-1} yr^{-1})								
ASTRAP/1978[a]	3.03	11.50	20.84	18.29	9.89	21.57	28.13	50.95	21.43
ASTRAP/1980[b]	3.62	12.28	24.49	16.02	13.11	23.55	26.52	48.44	27.15
ASTRAP/1981[b]	3.38	13.84	28.67	17.44	12.74	24.66	28.63	48.32	25.55
	Deposition in Year 2000 (Kg S Ha^{-1} yr^{-1})								
ASTRAP/1978[a]	3.93	13.48	21.37	16.78	9.64	20.63	27.27	52.92	25.52
ASTRAP/1980[b]	4.42	13.34	24.00	15.05	12.30	22.00	25.47	50.14	31.39
ASTRAP/1981[b]	4.16	14.98	28.23	16.57	11.91	23.35	27.81	50.22	29.67

[a]Based on 2 months of meteorological data (January and July)
[b]Based on 12 months of meteorological data

DR1 = Boundary Waters
DR2 = Algoma
DR3 = Muskoka
DR4 = Quebec
DR5 = S. Nova Scotia
DR6 = Vermont/New Hampshire
DR7 = Adirondacks
DR8 = Pennsylvania
DR9 = Smokies

Table 7 Optimized Approaches to Keeping Wet Sulfate Deposition Below 20 (30) Kg Ha^{-1} yr^{-1} in All Sensitive Regions, Modeled with ASTRAP Using 1980 Meteorological Data

	Annual Cost ($ billion)	Emission Reduction (MM tpy)	Reduction in Deposition (Kg S Ha^{-1} yr^{-1})								
			DR1	DR2	DR3	DR4	DR5	DR6	DR7	DR8	DR9
S.768	4.24	8.98	1.13	4.44	6.16	3.18	2.51	4.42	5.92	17.38	12.10
S.768 (cost opt.)	1.87	9.05	0.84	3.83	6.01	3.38	3.06	5.01	6.62	19.40	11.07
S.769	12.85	14.94	1.67	6.67	9.79	5.31	4.63	7.68	10.14	28.92	19.81
S.769 (cost opt.)	6.00	14.97	1.43	6.00	9.78	5.79	5.71	9.00	11.60	30.87	19.45
20 Kg Loading	11.91	16.16	1.52	7.04	12.07	6.94	5.79	10.57	14.23	40.73	18.52
(20 Kg Targets)[a]	-	-	0	1.68	12.07	3.21	0	7.74	11.10	33.74	18.27
30 Kg Loading	2.49	9.39	0.66	3.55	6.53	3.64	2.94	5.19	7.34	25.61	14.77
(30 Kg Targets)[a]	-	-	0	0	6.10	0	0	0.64	3.92	25.57	11.71

[a] Indicates minimum reduction in total S deposition necessary, relative to projected 2000 deposition, to keep wet $SO_4^=$ deposition levels in each region below 20 (30) Kg Ha^{-1} yr^{-1}

DR1 = Boundary Waters
DR2 = Algoma
DR3 = Muskoka
DR4 = Quebec
DR5 = S. Nova Scotia
DR6 = Vermont/New Hampshire
DR7 = Adirondacks
DR8 = Pennsylvania
DR9 = Smokies

Table 8 The Bills S.768 and S.769 Evaluated with No Fuel Switching
Using ASTRAP with 1980 Meteorological Data

	Annual Cost ($ billion)	Emission Reduction (MM tpy)	Reduction in Deposition (% of 2000 Deposition)								
			DR1	DR2	DR3	DR4	DR5	DR6	DR7	DR8	DR9
With Fuel Switching											
S.768	4.24	8.98	26	33	26	21	20	20	23	35	39
S.768 (cost opt)	1.87	9.05	19	29	25	22	25	23	26	29	35
S.769	12.85	14.94	38	50	41	35	38	35	40	58	63
S.769 (cost opt)	6.00	14.97	32	45	41	38	46	41	46	62	62
Without Fuel Switching											
S.768	4.61	8.98	26	33	26	21	20	20	23	35	39
S.768 (cost opt)	2.26	9.01	18	28	25	23	26	24	27	41	36
S.769	13.62	14.94	38	50	41	35	37	35	40	58	63
S.769 (cost opt)	7.01	15.01	32	45	41	39	49	42	47	62	60

DR1 = Boundary Waters
DR2 = Algoma
DR3 = Muskoka
DR4 = Quebec
DR5 = S. Nova Scotia
DR6 = Vermont/New Hampshire
DR7 = Adirondacks
DR8 = Pennsylvania
DR9 = Smokies

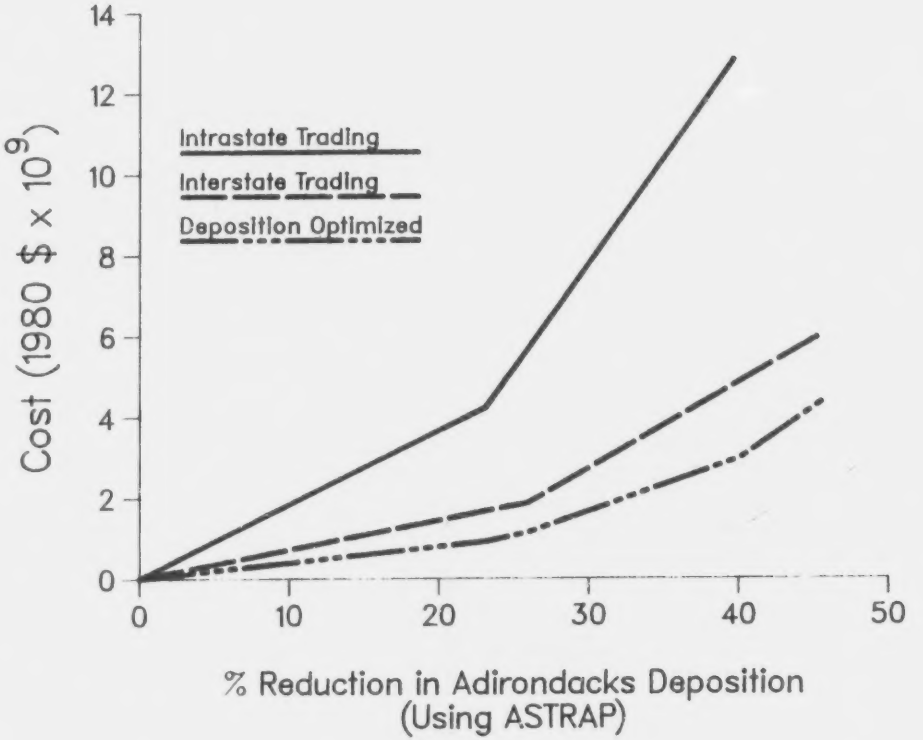

Fig. 1 Cost-Effectiveness of Alternative Strategies for Reduction of Sulfur Deposition in the Adirondacks

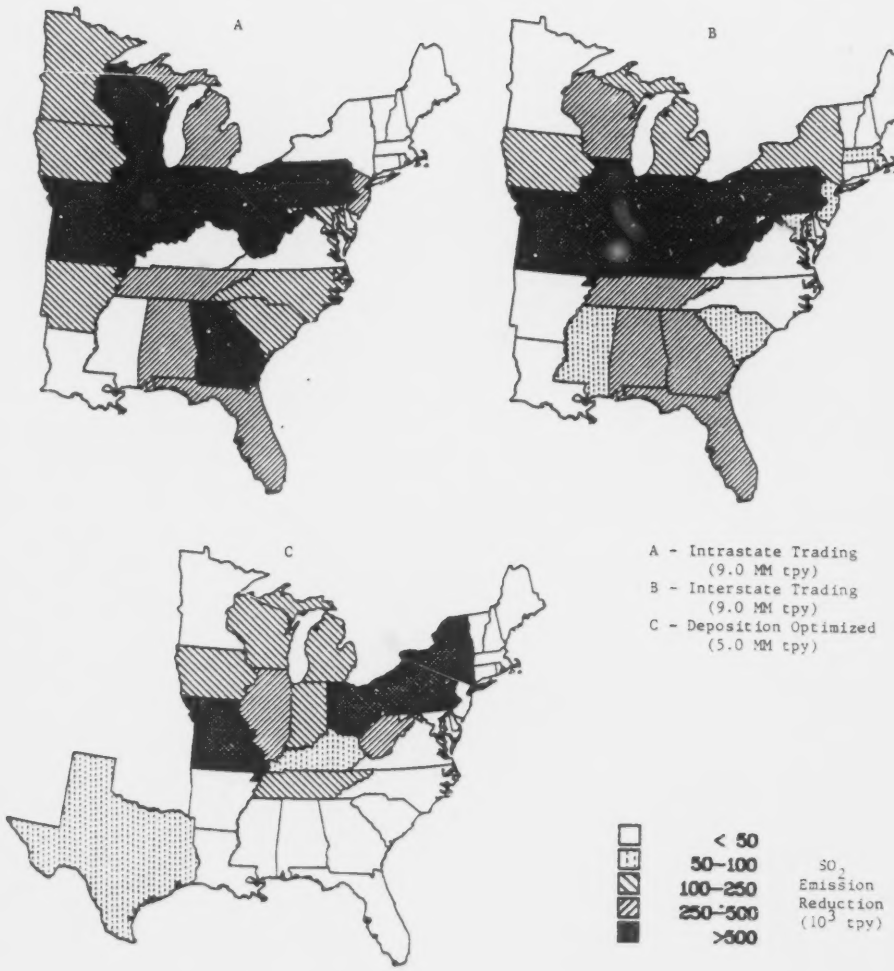

Fig. 2 State-Level Distribution of SO_2 Emission Reductions Required Under Alternative Versions of the Bill S.768

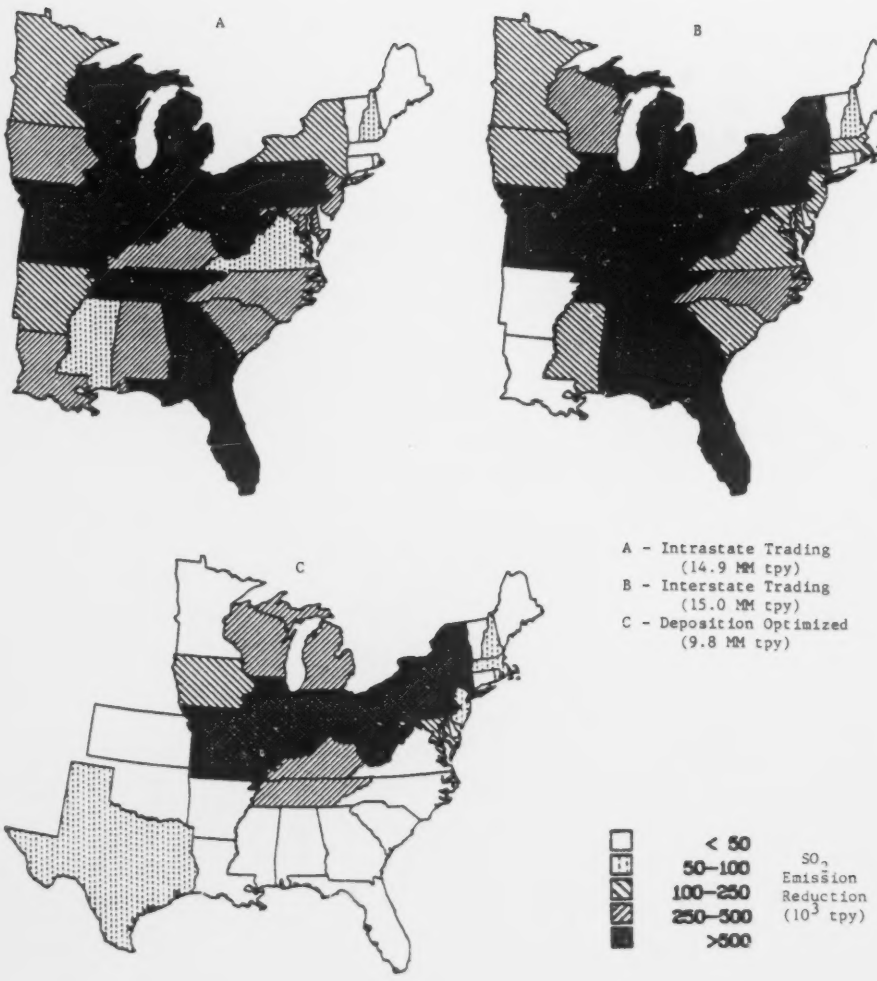

Fig. 3 State-Level Distribution of SO_2 Emission Reductions Required Under Alternative Versions of the Bill S.769

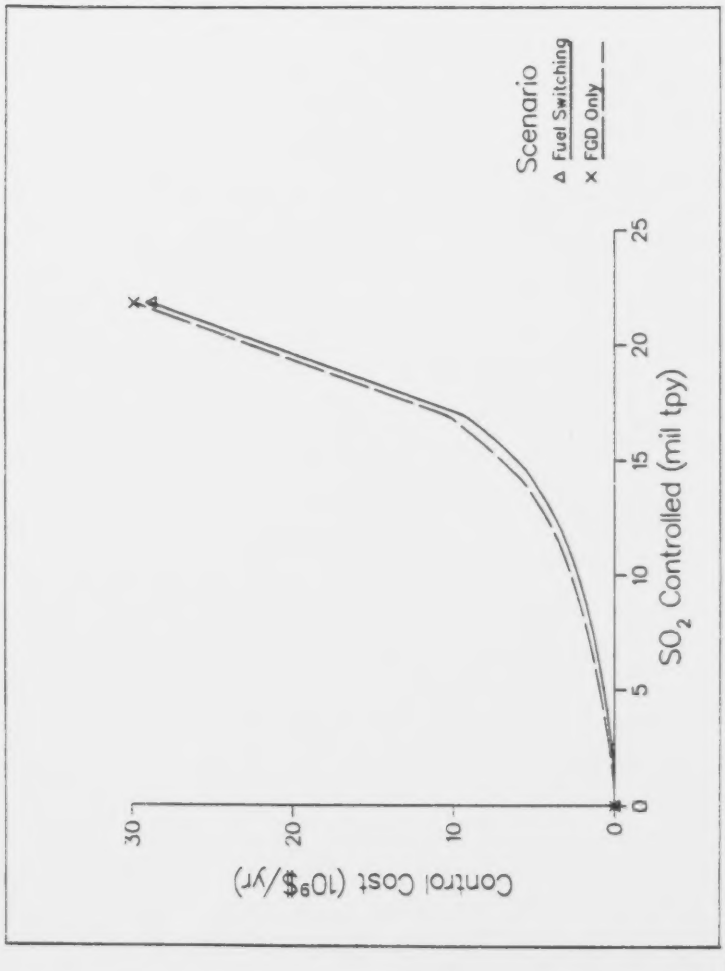

Fig. 4 Graph of Cost of SO_2 Control Versus SO_2 Emission Reduction (0-25 MM tpy) for a 31-State Optimization

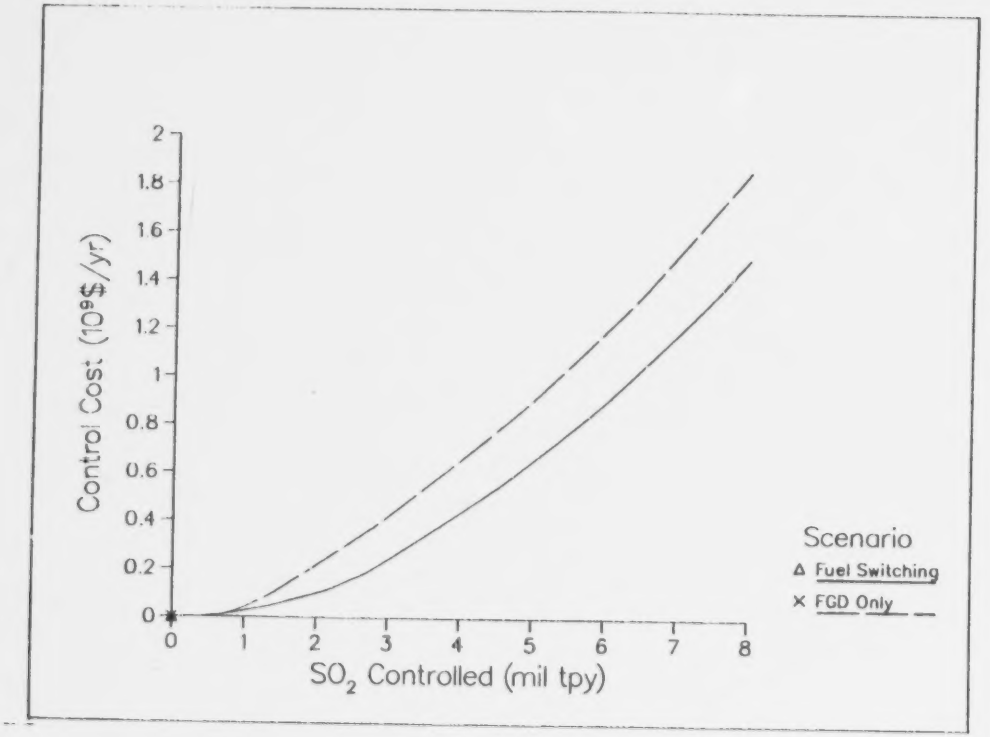

Fig. 5 Graph of Cost of SO_2 Control Versus SO_2 Emission Reduction (0-8 MM tpy) for a 31-State Optimization

APPENDIX A

CALCULATION OF STATE-LEVEL EMISSION REDUCTIONS

Tables A-1 and A-2 summarize the requirements of the two bills S.768 and S.769. The total emissions reductions required were allocated among the states according to the formulas contained in the bills. This was performed using a unit-specific inventory of utility power plants with information on current emissions, current emission rates, allowable emission rates, size of unit, and capacity utilization. New growth in SO_2 and NO_x emissions was obtained at the state level from the U.S.-Canadian MOI study.[10]

Table A-3 and Figure A-1 show the current SO_2 emissions from all sources in the 31-state eastern region. Table A-3 also shows the state-level distribution of emission reductions required by the bills, their cost-optimized alternatives, and their deposition-optimized alternatives. Maps of the reductions form Figs. 2 and 3 of the main text. These state-level emission reductions are utilized by the cost model (Appendix B) and the atmospheric transport models (Appendix C) to calculate cost increases and deposition reductions, respectively. It should be observed that, as specified by the bills, state allocations of the total reduction are determined on the basis of utility emissions, but the reductions themselves can be achieved by any source.

In order to determine the baseline deposition data for 1980 and 2000 (in the absence of control measures), a complete inventory of all sources in the United States and Canada is required. Table A-4 shows the emissions inventory used in this analysis, in a format appropriate for input to the atmospheric transfer matrices. See Tables C-1 and C-2 for identification of source regions, region numbers, and region name abbreviations.

Table A-1 Summary of S.768, The Stafford Bill/I

TITLE:	Adds Part E - "Interstate Transport and Acid Precursor Reduction" to Title I of the Clean Air Act.
IMPACT REGION:	31 States east of and bordering the Mississippi River, and the District of Columbia.
REGIONAL EMISSION INCREASE:	See SOURCE EMISSION INCREASE below. From 1/1/95, no SO_2 increase permitted; new sources meeting best-in-region BACT are exempt from offset requirements.
SOURCE EMISSION INCREASE:	No increase in actual SO_2 emissions as of 1/1/81, unless offset elsewhere in region; does not apply where the SIP limits all sources to 1.2 lb $SO_2/10^6$ Btu, or to coal conversion emissions >1.5 lb $SO_2/10^6$ Btu. No increase in actual NO_x emissions as of 1/1/81.
REDUCTION REQUIRED:	8 million tons/year SO_2 from 1980 actual emissions.
SCHEDULE FOR REDUCTION:	By 1/1/93, conventional retrofit technology in operation; by 1/1/95, innovative technology or replacement facility in operation.
STATE SHARE OF REDUCTION:	Governors allowed 18 months to develop allocation plan; failing agreement, state share equal to ratio of its utility (all) emissions (actual) in excess of 1.5 lb $SO_2/10^6$ Btu to region's total utility emissions (actual) in excess of 1.5 lb $SO_2/10^6$ Btu.
SOURCE OF REDUCTION:	Unspecified.
REDUCTION APPROACHES PERMITTED:	May be used if enforceable: 1) least emissions dispatch, 2) early retirement, 3) quantifiable energy conservation investment, and 4) regional trading.
NO_x CREDITS:	Not permitted.
NONCOMPLYING SOURCES:	Major sources not in SIP compliance in 1981 must be in compliance by 12/31/85 or be subject to 1.2 lb $SO_2/10^6$ Btu emission limitation.
STUDIES REQUIRED:	1) Additional funding for and reports from Interagency Task Force, 2) independent scientific review board established by NAS, 3) EPA and Treasury study of possible utility fee systems to finance emissions reductions, and 4) EPA NO_x inventory and control technology study.
REGULATORY AGENCY DEADLINES:	18 months from enactment: Gubernatorial allocation agreement. 42 months from enactment: SIP revision submittals or reductions. 6 months from submittal: EPA approval.
STATE DEFAULT:	Failure to reach gubernatorial agreement: see STATE SHARE OF REDUCTION above. Failure to submit or have approved SIP: 1.2 lb $SO_2/10^6$ Btu imposed on all utilities.

Table A-2 Summary of S.769, The Stafford Bill/II

TITLE: Acid Deposition and Sulfur Loadings Reduction Act, added as a new Part E to the Clean Air Act.

IMPACT REGION: 31 States east of and bordering the Mississippi River and the District of Columbia.

REGIONAL EMISSION INCREASE: See SOURCE EMISSION INCREASE below. Regional SO2 and NOx emissions from stationary sources may not increase over total actual emissions as of 1/1/81.

SOURCE EMISSION INCREASE: No increase in actual SO2 or NOx stationary source emissions over CY 80 actual emissions, unless offset by amount in excess of the increase through emission reductions not required by SIP. By 1/1/2000, all major stationary sources in region must be subject to an emission limitation at least as stringent as LAER (most stringent in any SIP for class of source, unless source demonstrates it is unachievable).

REDUCTION REQUIRED: 12 million tons/year SO2 from 1980 actual emissions.

SCHEDULE FOR REDUCTION: Phased; by 1/1/89, all reductions to be achieved through fuel switching; by 1/1/93, 5/6 of each State's total reduction to be achieved; by 1/1/98, total reduction to be achieved.

STATE SHARE OF REDUCTION: State share equal to ratio of its actual utility emissions in excess of 1.2 lb SO2/10(6) Btu to region's total utility actual emissions in excess of 1.2 lb SO2/10(6) Btu. Two or more States may reallot their total among themselves.

SOURCE OF REDUCTION: Unspecified.

REDUCTION APPROACHES PERMITTED: May be used if enforceable: (1) least emissions dispatch, (2) early retirement, (3) quantifiable energy conservation investment, (4) regional trading, and (5) precombustion cleaning of fuels.

NOx CREDITS: Allowed on basis of two units by weight of NOx for each SO2 unit.

STUDIES REQUIRED: Two-year study of long-range transport in continental U.S. outside acid deposition impact region.

REGULATORY AGENCY DEADLINES: 2 years from enactment: State submittal of reduction SIP requirements. 6 months from submittal: EPA approval.

STATE DEFAULT: All non-NSPS fossil-fuel powerplants, which emitted 1.2 lb SO2/10(6) Btu or greater in 1980, must comply with a thirty-day average, entity-wide, of 1.2 lb SO2/10(6) Btu for all such facilities owned or operated by the same entity.

A-5

Table A-3 State-Level Distribution of Emission Reductions Required Under S.768 and S.769 and Their Optimized Alternatives

	1980 Emissions (10^3 tpy)[a]	SO_2 Emission Reduction (10^3 English short tons per year					
		S.768	S.768 (cost opt)	S.768[b] (dep opt)	S.769	S.769 (cost opt)	S.769[b] (dep opt)
AL	757	291	263	0	483	502	3
AR	101	153	6	0	200	42	0
CT	72	41	6	6	49	25	6
DC	15	0	1	1	2	5	1
DE	109	-46	5	5	-26	40	21
FL	1093	355	354	0	642	663	0
GA	838	599	341	1	890	628	1
IA	328	220	142	115	284	235	138
IL	1468	985	851	370	1375	1099	851
IN	2004	553	1227	207	993	1506	1236
KY	1118	14	573	54	344	898	444
LA	304	0	12	0	465	49	0
MA	344	-91	80	9	-24	212	82
MD	337	185	94	14	281	195	157
ME	95	2	10	0	21	42	10
MI	905	363	198	159	563	528	473
MN	260	101	32	5	163	155	11
MO	1298	826	942	643	1142	1121	670
MS	284	42	80	7	58	137	7
NC	601	246	30	3	409	322	30
NH	93	40	46	46	69	73	70
NJ	279	257	74	45	346	109	86
NY	942	25	154	504	169	520	654
OH	2641	1262	1212	1001	2049	2042	2035
PA	2018	591	859	996	1036	1286	1286
RI	15	8	1	1	9	4	1
SC	325	241	74	0	329	227	6
TN	1074	396	475	177	665	874	391
VA	360	32	44	11	83	149	44
VT	7	7	1	1	8	2	2
WI	636	711	297	172	927	425	290
WV	1085	576	571	407	930	851	752
Total	21806	8985	9055	5013[c]	14934	14966	9829[d]

[a] From Ref. 10
[b] Using ASTRAP with 1980 meteorology
[c] Including a contribution of 53 x 10^3 tpy from Texas
[d] Including contributions of 53 x 10^3 tpy from Texas and 18 x 10^3 tpy from Kansas

Table A-4 Emissions of Sulfur Dioxide in the United States and Canada Used in the Atmospheric Transfer Matrices[a]

Source Region Number	SO_2 Emissions in 1980 (10^3 metric tons)	SO_2 Emissions in 2000 (10^3 metric tons)	Source Region Name
10	464	708	NMN
11	26	40	SMN
12	25	20	NWO
13	180	145	NEO
14	940	756	SUD
15	651	523	SWO
16	35	28	SEO
17	467	358	SLV
18	560	428	NQU
19	131	100	GBY
20	215	144	NBK
21	225	154	NSP
22	61	14	NFL
23	597	876	ASK
24	193	215	BCA
50	2401	2415	OHO
51	1334	1674	ILL
52	1835	1898	PEN
53	1822	1466	IND
54	1017	789	KEN
55	823	1010	MCH
56	977	997	TEN
57	1180	1227	MSU
58	986	1122	WVR
59	857	741	NYK
60	688	792	ALA
61	876	1394	WIO
62	236	291	MIN
63	874	1123	VNC
64	993	1093	FLD
65	1058	1465	GSC
66	673	962	MDJ
67	627	1232	ALM
68	392	364	MCR
69	86	85	MAN
70	91	96	VNH
71	516	560	WNE
72	1833	2503	WSE
73	344	425	WNW
74	1507	1114	WSW

[a]See Tables C-1 and C-2 for identification of source region names and source region numbers.

A-7

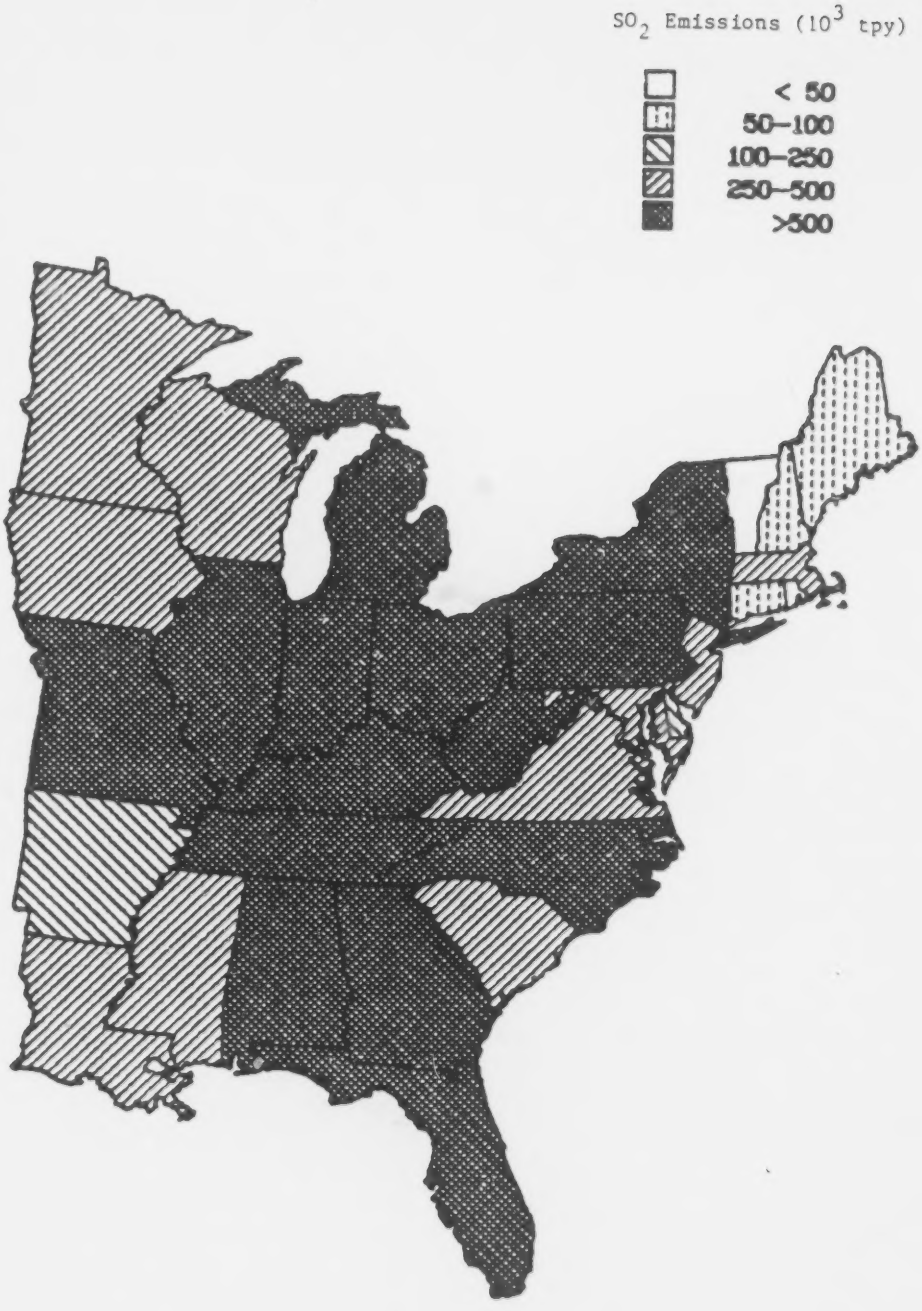

Fig. A-1 State-Level Distribution of 1980 SO_2 Emissions in the Eastern U.S.

APPENDIX B

DOCUMENTATION OF SO_2 REDUCTION POTENTIAL/CONTROL COST METHODOLOGY

Purpose

To estimate what reasonable potential exists for reducing SO_2 emissions in each state in the U.S., and to estimate the costs of achieving these reductions.

Baseline Data

The SO_2 emissions data from which the reduction potentials are estimated are contained in Tables B.2.1 and B.2.2 of the Work Group 3B Emissions, Costs and Engineering Assessment, performed under the U.S.-Canada Memorandum of Intent on Transboundary Air Pollution (Final Report, June 1982, pp 46-47).

This data set (Tables B-1 and B-2) contains 1980 SO_2 emissions for each state in the U.S. plus the District of Columbia, broken down into ten source categories. For the purposes of this analysis, only the contiguous U.S. was covered (omitting Alaska and Hawaii).

All emissions data are in metric tons per year, and all cost-effectiveness data are in dollars per metric ton of SO_2 removed. The cost calculations were all performed in metric tons; subsequently a conversion factor of 1.1 was used to convert emissions in metric tons to English short tons.

The Work Group 3B inventory shows 1980 total U.S. emissions of 24.09 million metric tons of SO_2, of which 15.76 million metric tons are emitted by electric utilities.

Reduction Potential in the Electric Utility Sector

This was determined through use of the AIRCOST model developed by E.H.

Pechan and Associates, based on the earlier AIRTEST model developed by Teknekron Research Incorporated. The 1980 inventory of power plants created by E.H. Pechan and Associates was used. AIRCOST was modified to permit the inclusion of fuel premiums in control costs.

The AIRCOST model was run for 16 different values of the SIP ceiling ranging between 12 $lb/10^6$ Btu and 0.1 $lb/10^6$ Btu. Units were not allowed to increase emissions above the level of actual 1980 emissions. This operation places increasingly stringent requirements on units until the final run in which all units emit at rates equal to or lower than 0.1 $lb/10^6$ Btu. The corresponding levelized costs in real 1980 dollars for each unit to meet the SIP ceilings were calculated. Least-cost control options were determined by computing marginal costs for each unit to reduce its emissions. However, some levels of emission reduction (i.e., SIP ceilings) may be clearly inefficient, in the sense that they would never be selected as part of a least-cost control strategy. That is, any least-cost control strategy would involve either higher or lower emissions for that unit.* A computer program was developed to eliminate inefficient emission reductions for a given unit. With these SIP ceilings eliminated, marginal costs were computed between the remaining emission levels. The emission reductions obtained were then sorted by state in order of increasing marginal cost.

Twelve marginal cost ranges were then selected, ranging from $0-300 per ton removed to >$5000 per ton removed. All utility emission reductions obtainable in each range, for each state, were then calculated. Table B-3 shows the form of these data. Significant SO_2 emissions remained even after

*Mathematically speaking, the set of emission/cost combinations which are worth considering lie on the convex hull of the set of all emission/cost combinations.

reducing all emission rates below 0.1 lb/10^6 Btu (OTHER column). These would be comprised of emissions remaining after application of FGD, emissions from combustion of very low sulfur coal, and emissions from oil combustion. It is not clear that these remaining emissions are "reasonably reducible." However, for the purposes of completeness they are included in the reduction potential tables at a nominal value of $4400/metric ton of SO_2 removed. Because the Work Group 3B utility inventory differs slightly from the E.H. Pechan utility inventory, an AIRCOST adjustment factor was used at the state level to scale utility emissions to the Work Group 3B inventory.

Weight-averaged marginal costs were calculated for each range, for each state, to correspond to the reduction potentials. Table B-4 lists these values in dollars/English short ton (adjusted later to dollars/metric ton). Mid-points of the range were written in this table if emissions were zero.

A maximum of 12 SO_2 emission reduction potentials and incremental control costs were therefore calculated for electric utilities in each state and passed on to the next phase of the analysis.

Reduction Potentials in the Non-Utility Sectors

These were treated in a less sophisticated way than the utility sector due to the absence of control cost models. Initially, each of the non-utility sectors listed in Tables B-1 and B-2 was examined individually to determine what SO_2 reductions could reasonably be achieved and at what incremental cost. A list of reduction potentials was drawn up, as shown in Table B-5. The reduction potentials are listed as a fraction of the Work Group 3B inventory amounts, and the marginal costs are listed in real 1980 dollars/English short ton removed. The data were derived from various sources, with costs converted to 1980 dollars using the CEN M&S Equipment Cost Index. Attachment

B-1 briefly describes how the non-utility potentials were determined and what assumptions were made about technology availability and performance.

All industrial cleanup opportunities were included that would cost less than $4400 per metric ton of SO_2 removed. Because of uncertainties in what reduction was reasonably achievable at high cost and what that cost would be, it was resolved to place an upper limit on marginal cost of $4400 per metric ton of SO_2 removed from non-utility sources. The balance of state non-utility emissions that could not be reduced at less than $4400 per metric ton was then, for purposes of completeness, determined to be achievable at that figure. This is discussed in more detail later. Reduction potentials from Table B-5 were then multiplied by state-level emissions in the appropriate source categories.

Total Reduction Potentials

The utility and non-utility reduction potentials were then merged and sorted by state in order of increasing marginal cost. Table B-6 shows an example for the State of Alabama. Costs (C) were determined for each reduction category by multiplying the reduction potential (A) by the weighted marginal cost (B). Cumulative costs (D) and emission reductions (E) were then determined, and average costs (F) for increasing levels of reduction were calculated. Finally, the cumulative reductions were expressed as a percentage of the state total.

Because of the assumptions made about the final two categories, other utility and other non-utility, 100% reduction is always potentially achievable. This table should be interpreted in the following way: 71% of the total SO_2 emissions in Alabama is determined to be "reasonably reducible" by application of available controls to major emitting sources, at an average

cost of $606/metric ton of SO_2 removed (marginal cost <$4400/ton). The remaining 29% of SO_2 emissions in Alabama will be very difficult to reduce and will involve untried control technologies or systems, or control of unconventional sources; we use a nominal cost of $4400/metric ton for control of this fraction of the state emissions.

Table B-7 shows the same table for Alabama when fuel switching in the utility sector is not permitted.

Calculation of the Costs of Specified Emission Reductions

One of the purposes of developing this methodology is to be able to determine least-cost methods of achieving reductions in SO_2 emissions at the state level, such as might be specified by an acid-rain control bill. A program was developed to read the state data as contained in Tables B-6 or B-7, move down the total tons column (E) until the required reduction is reached (with an appropriate interpolation scheme), and calculate the cost to achieve that level of reduction in the state. Marginal costs to remove the last ton of SO_2 (MC) and average costs (AC) are also determined. Finally, cumulative costs and emission reductions across all states are calculated. A typical analysis is presented in Table B-8. FRND4 is the state reduction amount.

Two problems typically occur in translating acid-rain control bills into state-level emission reductions: reductions greater than the state totals are required in some states (due to growth offsets), and "negative" reductions are required to some states (due to SIP compliance allowances). See Table A-3. In this analysis, it was assumed that additional reductions beyond state totals would be obtainable at $4400 per metric ton through interstate purchasing, and that "negative" reductions should be treated as emission "credits" at -$550/metric ton, available for sale to other states.

Two flags have been included in Table B-8. The flag "EM4400" identifies that portion of the emission reduction (in tons) which is achieved at the highest marginal cost of $4400/ton, both available within the state and unavailable within the state. That is to say, "EM4400" represents the difference between the required reduction and that amount of emissions which is determined to be "reasonably reducible."

The flag "EM FLAG" identifies that portion of the emission reduction (in tons) which is not available within the state. That is to say, "EM FLAG" represents the amount by which the required reduction exceeds the state total SO_2 emissions. Amounts in "EM4400" are always greater than the amounts in "EM FLAG." "EM FLAG" shows -1 if the reduction requirement is zero, and shows 0 if the reduction requirement is less than the state total emissions.

Calculation of Least-Cost Strategies for a Region

As a simple extension of the previous work, it is possible to estimate, least-cost ways to achieve various levels of emission reduction for a region, in the absence of state-specified reductions. A least-cost curve for a region is obtained simply by merging the data in Tables B-6 or B-7 for all states in the region, in order of increasing marginal cost. Cumulative emission reductions and costs can then be calculated and plotted. Figure 4 in the main text illustrates such a curve for the 31-state eastern region.

Warning

A series of tables and attachments are presented to illustrate the capabilities of the method and the format of the print-outs. They may not correspond to the final results quoted in the text. THE DATA PRESENTED IN THE ATTACHMENTS TO APPENDIX B ARE FOR ILLUSTRATIVE PURPOSES ONLY AND SHOULD NOT BE CITED.

Table B-1 1980 U.S. SO_2 Emissions Estimates
(10^3 metric tons/year)

State	Electric Utilities	Non-utility Combustion	Non-ferrous Smelters	Trans-portation	Other Sources	State Total
Alabama	492.7	77.7	1.5	12.1	103.5	688.3
Alaska	10.7	2.9	0.0	2.3	1.1	16.9
Arizona	79.4	7.9	699.7	8.5	20.6	816.1
Arkansas	24.1	28.7	12.9	9.2	17.2	92.1
California	70.7	50.6	0.0	91.0	192.7	405.0
Colorado	70.3	21.9	0.0	8.9	18.2	119.3
Connecticut	29.1	31.3	0.0	4.6	0.2	65.2
Delaware	47.6	23.9	0.0	1.9	25.8	99.2
Dist. of Columbia	4.2	7.7	0.0	0.9	0.6	13.4
Florida	658.5	87.9	0.0	38.6	208.3	993.3
Georgia	668.3	39.6	0.0	19.2	34.6	761.7
Hawaii	37.7	7.6	0.0	2.4	5.3	53.0
Idaho	0.0	10.3	17.2	3.7	11.3	42.5
Illinois	1 021.1	171.0	0.0	29.0	113.0	1 334.1
Indiana	1 396.7	263.1	4.5	19.8	137.4	1 821.5
Iowa	209.8	52.1	0.0	10.8	25.5	298.2
Kansas	136.2	10.2	0.0	13.6	42.1	202.1
Kentucky	914.0	60.3	6.3	13.0	23.1	1 016.7
Louisiana	22.5	69.4	5.4	27.8	150.9	276.0
Maine	14.8	59.0	0.0	3.2	9.0	86.0
Maryland	202.5	50.9	5.1	13.2	34.9	306.6
Massachusetts	249.9	52.5	0.0	9.3	0.8	312.5
Michigan	512.9	139.6	65.0	18.1	87.1	822.7
Minnesota	160.9	39.7	0.0	13.1	22.5	236.2
Mississippi	117.2	43.6	0.0	16.2	81.5	258.5
Missouri	1 034.7	49.9	26.8	17.2	51.8	1 180.4
Montana	21.2	22.9	78.0	5.0	21.5	148.6
Nebraska	44.9	3.9	0.0	9.2	10.0	68.0
Nevada	30.5	1.6	181.4	2.7	4.0	220.2
New Hampshire	73.0	9.1	0.0	1.4	0.8	84.3
New Jersey	100.0	67.6	0.0	22.9	62.8	253.3
New Mexico	76.8	2.0	57.4	8.5	99.0	243.7
New York	435.7	303.5	5.7	34.5	77.3	856.7
North Carolina	395.0	105.1	1.3	16.4	28.6	546.4
North Dakota	77.8	11.5	0.0	4.1	3.4	96.8
Ohio	1 970.1	281.7	6.8	31.3	111.2	2 401.1
Oklahoma	34.2	13.5	0.0	11.5	50.2	109.4
Oregon	3.0	22.8	3.6	13.5	11.6	54.5
Pennsylvania	1 330.1	230.2	2.7	33.9	237.6	1 834.5
Rhode Island	4.7	7.3	0.0	1.5	0.3	13.8
South Carolina	193.3	76.5	0.0	8.9	17.1	295.8
South Dakota	26.0	2.8	0.0	3.6	3.2	35.6
Tennessee	847.1	74.9	5.1	15.7	33.8	976.6
Texas	274.7	96.6	78.6	87.5	620.8	1 158.2
Utah	20.1	14.8	4.0	5.4	21.1	65.4
Vermont	0.5	4.4	0.0	1.3	0.0	6.2
Virginia	148.5	128.7	0.0	23.2	27.1	327.5
Washington	63.0	37.5	97.0	19.0	30.2	246.7
West Virginia	856.6	76.6	8.2	6.4	39.0	986.8
Wisconsin	440.6	96.9	0.0	13.2	27.5	578.2
Wyoming	106.7	26.8	0.0	6.2	27.0	166.7
TOTAL	15 760.5	3 178.8	1 374.4	795.8	2 982.7	24 092.2

Source: Work Group 3B Final Report (June 1982)

Table B-2 1980 U.S. SO_2 Emission Estimates for Major Categories Included in "Other Sources" (10^3 metric tons per year)

State	Petroleum Refining	Cement Plants	Nat. Gas Plants	Iron & Steel	H_2SO_4 Plants
Alabama	1.0	19.6	34.9	27.7	1.4
Alaska	1.2	0.0	0.0	0.0	0.0
Arizona	0.0	7.7	0.0	0.0	6.4
Arkansas	4.5	7.1	0.6	0.0	1.5
California	114.7	50.0	0.0	5.9	7.9
Colorado	1.7	10.5	0.0	3.1	0.0
Connecticut	0.0	0.0	0.0	0.0	0.0
Delaware	20.2	0.0	0.0	0.0	2.3
Dist. of Columbia	0.0	0.0	0.0	0.0	0.0
Florida	0.1	32.9	28.8	0.0	82.6
Georgia	0.4	9.3	0.0	0.0	3.1
Hawaii	2.4	2.4	0.0	0.0	0.2
Idaho	0.0	1.3	0.0	0.0	8.8
Illinois	68.6	18.7	0.0	13.8	6.7
Indiana	35.4	22.8	0.0	70.1	4.1
Iowa	0.0	21.6	0.0	0.0	3.0
Kansas	17.8	17.4	0.0	0.0	0.5
Kentucky	9.8	4.4	0.0	4.4	1.3
Louisiana	90.3	7.7	1.8	0.0	33.5
Maine	0.0	3.0	0.0	0.0	0.3
Maryland	0.8	15.3	0.0	15.0	1.6
Massachusetts	0.0	0.0	0.0	0.0	0.5
Michigan	7.5	42.2	1.2	21.8	0.4
Minnesota	16.0	0.0	0.0	0.0	0.0
Mississippi	14.4	4.5	43.0	0.0	6.5
Missouri	7.5	36.4	0.0	1.2	1.2
Montana	11.0	3.8	0.0	0.0	1.4
Nebraska	0.3	3.8	0.0	0.0	0.0
Nevada	0.0	2.4	0.0	0.0	0.0
New Hampshire	0.3	0.0	0.0	0.0	0.0
New Jersey	31.9	0.0	0.0	0.0	6.5
New Mexico	2.6	1.8	85.3	0.0	3.1
New York	6.4	33.5	0.0	18.1	0.5
North Carolina	0.1	4.0	0.0	0.0	15.2
North Dakota	3.3	0.0	0.0	0.0	0.0
Ohio	26.4	16.5	0.0	54.6	3.2
Oklahoma	35.5	11.5	0.0	0.0	0.5
Oregon	0.2	3.5	0.0	0.0	0.0
Pennsylvania	45.6	56.3	0.0	110.1	1.9
Rhode Island	0.0	0.0	0.0	0.0	0.0
South Carolina	0.0	11.5	0.0	0.0	0.3
South Dakota	0.0	2.8	0.0	0.0	0.0
Tennessee	0.4	10.2	0.0	0.6	8.2
Texas	281.5	62.5	206.2	7.2	16.4
Utah	9.7	3.3	0.0	5.0	2.7
Vermont	0.0	0.0	0.0	0.0	0.0
Virginia	4.4	4.2	0.0	0.0	4.1
Washington	14.2	7.9	0.0	0.0	0.4
West Virginia	0.8	6.4	0.0	23.2	0.7
Wisconsin	1.3	2.3	0.0	1.2	0.0
Wyoming	18.3	2.0	5.4	0.0	0.5
TOTAL	998.4	594.8	407.1	382.9	239.1

Source: Work Group 3B Final Report (June 1982)

Table B-3 Utility Emission Reductions as a Function of Marginal Cost

STATE	EM0_3	EM3_4	EM4_5	EM5_6	EM6_7	EM7_8	EM8_9	EM9_10	EM10_11	EM11_12	EM12_20	OVR20E	OTHER	BASEMS
AL	2.3	100.2	18.5	24.6	76.8	19.6	13.6	31.1	23.6	10.1	32.9	8.4	119.5	483.3
AR	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	1.3	11.4	12.5
AZ	2.6	2.7	0.0	0.0	0.0	0.0	0.0	28.2	0.0	2.1	4.3	1.8	38.7	100.5
CA	0	0	0	0	0	0	0	0	0	0	0	0	34.6	34.6
CO	0.0	0.0	0.0	1.3	0.0	0.0	0.0	0.0	0.0	2.9	7.7	10.1	30.4	75.4
CT	0	0	0	0	0	0	0	0	0	0	0	0	29.0	29.0
DC	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	1.8	2.3	4.1
DE	0.0	0.3	0.0	0.0	0.0	0.0	0.0	0.0	2.5	3.6	3.0	2.6	17.2	29.2
FL	96.7	99.6	84.2	103.1	45.3	16.2	25.1	14.7	5.1	37.5	33.8	22.7	106.6	704.6
GA	0.7	0.0	142.9	78.1	78.8	72.2	46.8	41.4	34.0	2.8	21.1	14.3	127.7	660.8
IA	100.8	1.1	0.6	2.8	10.6	9.5	0.0	10.9	10.1	1.0	9.7	12.8	30.0	222.9
ID	0	0	0	0	0	0	0	0	0	0	0	0	0.0	0.0
IL	311.9	153.5	158.8	115.8	0.0	0.0	12.2	0.0	0.5	23.3	46.5	30.3	199.4	1006.2
IN	147.5	295.0	337.3	130.9	132.1	60.2	8.2	37.5	29.0	20.1	23.0	52.6	172.1	1445.4
KS	9.0	1.5	5.9	6.3	10.5	0.0	0.0	1.9	0.0	0.0	0.0	12.4	45.0	93.3
KY	31.5	17.4	104.7	127.6	80.0	13.5	12.0	10.8	3.2	23.9	25.4	45.1	174.3	673.3
LA	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	7.1	0.0	0.0	7.2	14.3
MA	0.0	0.0	0.0	0.0	20.5	14.1	1.0	0.0	20.7	4.5	11.3	2.8	80.5	155.1
MD	0.0	4.3	0.0	46.2	25.1	4.2	21.6	0.0	12.6	9.5	9.1	21.8	49.8	207.5
ME	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.9	0.0	0.0	0.3	1.2
MI	1.7	0.0	17.8	108.0	29.1	7.8	0.0	0.0	71.5	34.5	9.3	21.4	144.7	565.6
MN	5.3	6.6	0.0	15.5	0.0	16.5	0.0	0.0	32.8	6.5	7.0	6.4	53.8	194.8
MO	297.9	17.6	189.7	58.7	0.0	23.5	0.0	0.0	30.4	0.0	15.9	22.5	122.7	841.1
MS	3.4	0.0	0.0	0.0	52.7	8.3	0.0	0.0	0.0	0.0	7.0	2.7	25.0	119.6
MT	0.0	4.4	0.0	0.0	0.0	0.0	0.0	0.0	3.4	0.0	1.2	0.0	14.7	38.6
NB	11.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.3	8.0	6.7	22.1	49.0
NC	3.2	0.0	0.0	8.1	0.0	30.5	80.7	35.3	69.7	14.0	14.8	21.0	144.6	421.4
ND	5.6	0.0	0.0	0.0	7.8	14.6	7.2	12.0	4.7	5.3	1.4	1.2	34.2	94.0
NH	16.0	0.0	0.0	23.8	0.0	8.7	0.0	0.0	0.0	10.0	0.0	0.0	13.9	72.3
NJ	20.4	7.9	0.0	0.0	19.2	12.4	0.0	0.0	3.7	0.0	2.0	2.7	26.2	94.5
NM	4.4	0.0	0.0	0.0	0.0	0.0	6.4	0.0	4.5	0.0	0.0	0.0	30.6	86.0
NV	0.0	0.0	0.0	1.3	1.1	0.0	0.0	1.0	7.4	0.0	0.0	0.0	23.4	40.2
NY	1.4	0.0	0.0	31.1	50.7	74.5	34.4	13.9	37.7	17.5	27.1	13.3	121.4	426.9
OH	380.5	255.3	35.1	122.6	170.9	139.0	136.6	60.8	93.6	34.4	82.0	26.0	279.1	1617.8
OK	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.3	0.3	23.8	40.4
OR	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.5	2.0	2.5
PA	208.9	0.0	172.9	173.5	173.7	70.8	31.4	93.0	55.7	20.5	1.7	2.4	279.0	1303.7
RI	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	1.2	0.9	2.0	4.6
SC	2.5	4.1	0.0	0.0	19.5	38.4	34.5	5.4	27.7	12.7	16.2	13.7	32.8	227.6
SD	0.0	0.0	0.0	0.0	10.7	0.0	7.6	0.0	0.0	0.0	0.0	0.0	7.6	25.9
TN	114.2	50.6	70.7	93.7	0.0	13.1	50.1	14.8	60.2	0.0	28.5	9.9	120.0	599.8
TX	29.7	4.9	0.0	0.0	0.0	32.7	31.9	0.3	82.7	0.0	2.6	0.4	168.2	358.8
UT	0.0	0.9	0.0	0.0	0.0	7.3	0.0	0.7	0.0	0.0	0.0	0.8	28.5	43.9
VA	4.6	0.0	0.0	3.5	0.6	8.0	8.0	19.9	14.5	9.4	24.3	10.0	57.1	133.0
VT	0	0	0	0	0	0	0	0	0	0	0	0	0.0	0.0
WA	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	11.0	9.1	0.7	0.0	20.4	41.1
WI	104.6	9.7	19.4	32.0	4.5	4.5	11.8	9.5	4.0	3.1	3.8	21.8	80.0	296.7
WV	91.8	218.5	19.2	57.2	49.2	119.1	89.5	30.7	28.1	7.6	20.3	35.2	173.0	949.3
WY	0.0	2.2	0.0	0.0	0.0	0.0	0.0	5.6	4.9	9.2	0.0	2.8	50.5	70.9

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Table B-4 Weight-Averaged Marginal Costs for Utility Emission Reductions in Each Marginal Cost Range

STATE	MC0_3	MC3_6	MC6_9	MC9_12	MC12_15	MC15_18	MC18_21	MC21_24	MC24_27	MC27_30	OVR30C	OTHERC
AL	190.4	388.1	459.4	581.2	635.9	731.0	873.0	965.1	1027.1	1374.0	1674.3	2813.2	5000
AR	150.0	350.0	450.0	550.0	650.0	750.0	850.0	950.0	1100.0	1350.0	1750.0	4203.7	5000
AZ	93.3	375.4	450.0	550.0	650.0	750.0	850.0	950.0	1100.0	1431.1	1708.9	2043.2	5000
CA	0	0	0	0	0	0	0	0	0	0	0	0	5000
CO	150.0	350.0	450.0	508.2	650.0	750.0	850.0	950.0	1100.0	1350.7	1838.8	3021.3	5000
CT	0	0	0	0	0	0	0	0	0	0	0	0	5000
DC	150.0	350.0	450.0	550.0	650.0	750.0	850.0	950.0	1100.0	1350.0	1750.0	4196.1	5000
DE	150.0	388.4	450.0	550.0	650.0	750.0	850.0	950.0	1149.1	1443.4	1901.0	3231.0	5000
FL	242.3	382.9	429.9	552.3	634.8	767.2	879.7	939.8	1090.2	1350.4	1769.7	2523.1	5000
GA	0.0	350.0	471.9	547.0	670.6	750.4	853.2	929.0	1138.2	1362.0	1717.7	3040.2	5000
IA	78.0	377.3	437.2	538.3	632.6	741.9	850.0	923.1	1102.7	1343.9	1880.6	4071.0	5000
ID	0	0	0	0	0	0	0	0	0	0	0	0	5000
IL	193.3	384.6	429.8	524.3	650.0	750.0	809.5	950.0	1088.8	1433.8	1752.7	3067.9	5000
IN	83.6	369.0	454.9	546.2	652.2	737.2	808.4	917.0	1081.0	1250.6	1776.8	3287.0	5000
KS	58.7	379.5	459.2	569.4	617.5	750.0	850.0	940.0	1100.0	1350.0	1750.0	3129.7	5000
KY	11.3	370.7	442.8	545.9	663.8	753.5	842.1	944.7	1081.2	1293.6	1709.1	3241.3	5000
LA	150.0	350.0	450.0	550.0	650.0	750.0	850.0	950.0	1100.0	1427.1	1750.0	3000.0	5000
MA	150.0	350.0	450.0	550.0	679.4	702.9	808.[illegible]	950.0	1100.0	1321.1	1749.8	2394.1	5000
MD	0.0	390.7	450.0	538.6	653.8	748.0	[illegible]	774.0	1082.7	1380.2	1642.4	2670.1	5000
ME	150.0	350.0	450.0	550.0	650.0	750.0	850.0	950.0	1100.0	1218.3	1750.0	3000.0	5000
MI	216.3	350.0	481.0	522.8	655.5	7[illegible]	[illegible]	930.2	1087.0	1298.1	1777.0	3342.7	5000
MN	121.4	326.9	450.0	594.5	650.0	75[illegible]	[illegible]	950.3	1161.0	1310.6	1718.9	4809.8	5000
MO	167.4	390.1	449.2	560.6	650.0	711.[illegible]	[illegible]	950.0	1070.9	1350.0	1750.2	3220.9	5000
MS	0.3	350.0	450.0	550.0	639.2	722.[illegible]	[illegible]	939.3	1100.0	1350.0	1972.8	2318.1	5000
MT	130.0	329.6	450.0	550.0	650.0	750.0	859.1	944.7	1070.8	1370.0	1681.9	3000.0	5000
NB	277.1	350.0	450.0	550.0	650.0	750.0	850.0	983.8	1100.0	1250.0	1750.0	3107.5	5000
NC	123.9	350.0	450.0	582.5	650.0	741.9	858.8	955.1	1071.0	1323.5	1770.1	2601.7	5000
ND	210.8	350.0	450.0	550.0	608.9	781.9	832.8	918.3	1049.8	1361.5	1657.4	2120.1	5000
NH	140.8	350.0	450.0	552.4	650.0	767.0	850.0	950.0	1100.0	1369.8	1750.0	3000.0	5000
NJ	106.2	218.0	450.0	550.0	624.0	740.8	850.0	950.0	1083.9	1350.0	1531.4	2343.0	5000
NM	270.9	350.0	450.0	550.0	650.0	750.0	859.3	950.0	1051.2	1350.0	1750.0	00.0	5000
NV	150.0	350.0	450.0	503.4	626.8	750.0	850.0	902.5	1070.3	1248.2	1750.0	00.0	5000
NY	0.7	350.0	450.0	577.0	651.1	754.0	835.5	917.5	1088.2	1334.2	1769.8	3076.0	5000
OH	112.9	347.1	444.0	559.7	652.1	757.9	841.5	943.1	1100.7	1342.5	1736.2	3037.2	5000
OK	150.0	350.0	450.0	550.0	650.0	750.0	850.0	950.0	1100.0	1350.0	1802.4	3101.5	5000
OR	150.0	350.0	450.0	550.0	650.0	750.0	850.0	950.0	1100.0	1350.0	1750.0	12319.0	5000
PA	102.8	350.0	478.9	544.9	627.9	765.7	840.3	943.0	1056.8	1277.2	1867.3	3143.0	5000
RI	150.0	350.0	450.0	550.0	650.0	750.0	850.0	950.0	1100.0	1350.0	1914.8	3087.1	5000
SC	132.3	311.5	450.0	550.0	628.1	738.1	844.8	936.9	1051.1	1349.0	1657.0	2507.0	5000
SD	150.0	350.0	450.0	550.0	668.4	750.0	807.0	950.0	1100.0	1350.0	1750.0	3000.0	5000
TN	154.0	374.4	424.6	548.8	650.0	760.7	857.3	979.0	1079.8	1350.0	1770.4	2783.8	5000
TX	38.4	323.5	450.0	550.0	650.0	750.3	811.8	950.0	1087.5	1350.0	1672.8	3298.9	5000
UT	150.0	329.1	450.0	550.0	650.0	734.1	850.0	943.0	1100.0	1350.0	1750.0	2490.0	5000
VA	0.2	350.0	450.0	529.1	665.8	750.0	832.0	941.0	1103.0	1333.2	1783.6	3336.3	5000
VT	0	0	0	0	0	0	0	0	0	0	0	0	5000
WA	130.0	350.0	450.0	550.0	650.0	750.0	850.0	950.0	1117.6	1381.5	1695.1	3000.0	5000
WI	180.8	319.7	469.2	576.8	688.4	790.2	886.3	982.7	1087.0	1441.0	1829.5	3706.7	5000
WV	154.4	359.0	429.4	547.9	647.5	737.0	858.2	969.4	1085.8	1498.9	1787.7	2833.5	5000
WY	150.0	341.1	450.0	550.0	650.0	750.0	850.0	980.8	1019.3	1240.0	1750.0	6140.6	5000

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Table B-5 Non-Utility SO_2 Emission Reduction Potentials

1.	0.16x	at	$200/ton	IBEW	Industrial Boilers/East/Wash
2.	0.11x	at	$790/ton	IBES	Industrial Boilers/East/Switch
3.	0.21x	at	$1450/ton	IBEF	Industrial Boilers/East/FGD
4.	0.12x	at	$880/ton	IBE[illegible]	Industrial Boilers/East/Oil
5.	0.11x	at	$2650/to[illegible]	[illegible]F	Industrial Boilers/West/FGD
6.	0.12x	at	$880/ton	IBW[illegible]	Industrial Boilers/West/Oil
7.	0.66x	at	$675/ton	SMLT	Smelters
8.	0.41x	at	$1170/ton	CMNT	Cement Industry
9.	0.23x	at	$2130/ton	PROS	Oil Switch in Refinery Process Heaters
10.	0.30x	at	$3300/ton	PRTG	Pet. Ref. Tail Gas Clean-Up

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x = Emissions from W-G 3B Inventory

Marginal Costs in 1980 $ per English short ton

Table B-6 State SO_2 Emission Reduction Potential Using a Least-Cost Methodology
(With Fuel Switching Permitted for Electric Utilities)

STATE = AL
REGION = E
SO2 EMISSIONS : STATE TOTAL = 689.9 (1000 METRIC TONS)
UTILITY EMISSIONS = 498.0 (1000 METRIC TONS)
AIRCOST ADJUSTMENT FACTOR = 1.122

SOURCE	TONS REM. (A)	MC (B)	COST (C)	TC (D)	TOTAL TONS (E)	AC (F)	PCT (G)
UTIL	2.5	111.	0.3	0.3	2.5	111.	0.4
IBEW	12.5	220.	2.7	3.0	15.0	201.	2.2
UTIL	102.2	220.	23.1	26.1	117.2	223.	17.0
UTIL	18.9	267.	5.0	31.1	136.1	229.	19.7
UTIL	25.1	330.	8.5	39.6	161.2	246.	23.4
UTIL	78.3	370.	29.0	68.6	239.5	286.	34.7
UTIL	20.0	437.	8.7	77.3	259.5	298.	37.6
UTIL	13.9	506.	7.0	84.4	273.4	309.	39.6
UTIL	31.7	561.	17.8	102.2	305.1	335.	44.2
UTIL	37.9	619.	23.5	125.7	343.0	366.	49.7
SMLT	1.3	749.	1.0	126.7	344.3	368.	49.9
UTIL	59.7	822.	49.1	175.7	404.0	435.	58.6
IBEE	8.6	869.	7.5	183.2	412.6	444.	59.8
IBED	9.4	968.	9.1	192.2	421.9	456.	61.2
UTIL	9.9	1004.	9.9	202.2	431.8	468.	62.6
CMNT	8.2	1207.	10.6	212.7	440.0	483.	63.8
IBEF	16.4	1595.	26.1	238.8	456.4	523.	66.2
UTIL	32.5	1739.	56.4	295.2	488.9	604.	70.9
PROS	0.2	2248.	0.5	295.8	489.2	605.	70.9
PRTG	0.3	3630.	1.1	296.8	489.5	606.	70.9
UTIL	60.4	4400.	265.7	562.5	549.8	1023.	79.7
OTHR	140.0	4400.	616.2	1178.7	689.9	1709.	100.0

(A) 1000 METRIC TONS OF SO2 REMOVED
(B) MARGINAL COST (1980 $/METRIC TON)
(C) COST (MILLION 1980 $), (A)*(B)
(D) TOTAL COST (MILLION 1980 $), (CUMMULATIVE (C))
(E) SO2 REMOVED (1000 METRIC TONS), (CUMMULATIVE (A))
(F) AVERAGE COST (1980 $/METRIC TON) (D)/(E)
(G) PERCENT SO_2 REDUCTION OF STATE TOTAL, (E)/STATE EMISSION

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Table B-7 State SO_2 Emission Reduction Potential Using a Least Cost Methodology
(with Fuel Switching Not Permitted for Electric Utilities)

STATE = AL
REGION = E
SO2 EMISSIONS : STATE TOTAL = 689.9 (1000 METRIC TONS)
UTILITY EMISSIONS = 493.0 (1000 METRIC TONS)
AIRCOST ADJUSTMENT FACTOR = 1.095

SOURCE	TONS REM. (A)	MC (B)	COST (C)	TC (D)	TOTAL TONS (E)	AC (F)	PCT (G)
IBEW	12.5	220.	2.7	2.7	12.5	220.	1.8
UTIL	66.0	329.	21.3	24.1	78.5	307.	11.4
UTIL	68.5	368.	25.2	49.3	147.0	335.	21.3
UTIL	69.4	437.	30.3	79.6	216.4	368.	31.4
UTIL	33.0	507.	16.7	96.3	249.4	386.	36.1
UTIL	49.0	561.	27.5	123.8	298.4	415.	43.2
UTIL	41.2	615.	25.	148.8	339.6	439.	49.2
SMLT	1.3	742.	1.0	149.8	340.9	440.	49.4
UTIL	19.1	765.	14.6	164.4	360.0	458.	52.2
IBBS	8.6	869.	7.5	172.2	368.6	467.	53.4
IBEO	9.4	968.	9.1	181.3	378.0	480.	54.8
UTIL	30.2	1061.	32.0	213.3	408.1	523.	59.2
CMNT	8.2	1287.	10.6	223.9	416.3	538.	60.3
IBEF	16.4	1595.	26.1	250.0	432.7	578.	62.7
UTIL	60.2	1940.	116.9	366.9	493.0	744.	71.5
PROS	0.2	2343.	0.5	367.4	493.2	745.	71.5
PRTG	0.3	3630.	1.1	368.5	493.5	747.	71.5
UTIL	56.6	4400.	248.0	616.5	549.8	1121.	79.7
OTHR	140.0	4400.	616.2	1232.7	689.9	1787.	100.0

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(A) 1000 METRIC TONS OF SO2 REMOVED
(B) MARGINAL COST (1980 $/METRIC TON)
(C) COST (MILLION 1980 $), (A)*(B)
(D) TOTAL COST (MILLION 1980 $), (CUMMULATIVE (C))
(E) SO2 REMOVED (1000 METRIC TONS), (CUMMULATIVE (A))
(F) AVERAGE COST (1980 $/METRIC TON) (D)/(E)
(G) PERCENT SO2 REDUCTION OF STATE TOTAL, (E)/STATE EMISSION

Table B-8 Sample Cost Analysis of An Acid Deposition Control Bill

LEAST COSTS FOR 8MM TPY SO2 REDN--ANL ASTRAP DEP. 1981
CALCULATED LIKE S768 (5 GROWTH)
INTRASTATE TRADING

OBS	STATE	SOURCE	MC	AC	CUM_COST	ER_FLAG	ST_COST	TOT_GOAL	ER4400	FRND4
1	AL	UTIL	508	301.59	79.67	0.0000	79.67	264.16	0.000	264.16
2	AR	OTHR	4400	3682.49	593.29	47.4766	513.62	403.63	91.677	139.48
3	CT	UTIL	4400	1784.21	659.22	0.0000	65.93	440.59	6.552	36.95
4	DC	IBEW	550	550.00	659.22	-1.0000	-0.00	440.59	0.000	-0.00
5	DE	IBEW	550	550.00	636.08	-1.0000	-23.14	398.51	0.000	-42.07
6	FL	UTIL	446	261.50	720.52	0.0000	84.44	721.42	0.000	322.91
7	GA	UTIL	942	426.62	952.75	0.0000	232.24	1265.79	0.000	544.36
8	IA	CMNT	1287	349.35	1022.48	0.0000	69.73	1465.38	0.000	199.59
9	IL	IBEW	968	266.40	1261.19	0.0000	238.71	2361.18	0.000	895.80
10	IN	IBEW	220	149.54	1336.34	0.0000	75.15	2863.75	0.000	502.58
11	KY	UTIL	14	14.00	1336.51	0.0000	0.17	2876.06	0.000	12.30
12	LA	IBEW	550	550.00	1336.51	-1.0000	-0.00	2876.06	0.000	-0.00
13	MA	IBEW	550	550.00	1290.96	-1.0000	-45.55	2793.23	0.000	-82.83
14	MD	UTIL	984	519.72	1378.53	0.0000	87.57	2961.73	0.000	168.50
15	ME	IBEW	220	220.00	1378.89	-1.0000	0.36	2963.36	0.000	1.63
16	MI	UTIL	603	430.49	1521.03	0.0000	142.14	3293.54	0.000	330.18
17	MN	UTIL	675	445.81	1561.81	0.0000	40.78	3385.01	0.000	91.46
18	MO	UTIL	262	121.06	1652.87	0.0000	90.86	4135.58	0.000	750.57
19	MS	UTIL	372	284.61	1663.54	0.0000	10.67	4173.75	0.000	38.18
20	NC	UTIL	623	502.85	1775.95	0.0000	112.41	4397.30	0.000	223.54
21	ND	UTIL	421	207.38	1783.52	0.0000	7.57	4433.82	0.000	36.52
22	NJ	OTHR	4400	2263.86	2312.23	0.0000	528.71	4667.36	86.143	233.54
23	NY	IBEW	220	206.35	2316.89	0.0000	4.66	4689.92	0.000	22.56
24	OH	UTIL	441	208.16	2555.63	0.0000	238.75	5836.88	0.000	1146.96
25	PA	UTIL	317	184.09	2654.52	0.0000	98.89	6374.04	0.000	537.16
26	RI	UTIL	1686	1179.77	2663.31	0.0000	8.79	6381.49	0.000	7.45
27	SC	IBEF	1595	715.95	2820.48	0.0000	157.16	6601.01	0.000	219.52
28	TN	UTIL	319	174.31	2883.16	0.0000	62.69	6960.63	0.000	359.62
29	VA	IBEW	220	175.11	2888.28	0.0000	5.12	6989.86	0.000	29.23
30	VT	OTHR	4400	3230.97	2909.36	0.5226	21.07	6996.38	4.223	6.52
31	WI	OTHR	4400	1843.83	4100.83	68.1946	1191.47	7642.58	200.495	646.19
32	WV	UTIL	499	264.97	4239.56	0.0000	138.73	8166.14	0.000	523.57

NOTE: DATA SET WORK.A HAS 32 OBSERVATIONS AND 10 VARIABLES. 243 OBS/TRK.

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Attachment B-1 Derivation of Non-Utility SO_2 Emission Reduction Potentials and Costs

- Non-Utility Fuel Combustion

This sector was initially divided into industrial fuel combustion emissions (2.38×10^6 t/y) and residential/commercial fuel combustion emissions (0.8×10^6 t/y - W.G. 3B Report and NEDS Trends for 1980). The latter were carried forward to the Other Sources category at \$4400/ton SO_2 removed.

The industrial boiler (IB) emissions sources were further broken down into East/West and Coal/Oil Shares as follows: coal 70%, oil 30% (East); coal 17%, oil 83% (West), on the basis of EPA's 1980 Trends Report and ANL/EES-TM-54. As fractions of the Work Group 3B Inventory emissions (x), the controllable emissions are then: coal 0.53x, oil 0.23x (East); coal 0.13x, oil 0.62x (West).

The following control options were then considered:

East - Coal Washing (30% reduction at \$230/ton - Radian)
Fuel Switching (50% reduction at \$500/ton - ICF)
Scrub High-S Coal (90% reduction at \$1000/ton - ICF)
Oil Desulfurization (50% reduction at \$1000/ton - Radian)

West - Scrub Low-S Coal (85% reduction at \$3000/ton - ICF)
Oil Desulfurization (50% reduction* at \$1000/ton - Radian)

After the control options for coal in the East are converted into marginal costs and incremental emission reductions, the six control options IBEW, IBES, IBEF, IBEO, IBWF, and IBWO are obtained, as listed in Table B-5.

*But assume only 40% of the oil is amenable to desulfurization.

- Smelters

It has been estimated that 97% of uncontrolled SO_2 from smelters occurs in the West, and that 85% of SO_2 in the West is attributable to copper smelters (Mangeng and Mead, LASL Report LA-8268-MS, August 1980). Overall control of SO_2 emissions from existing copper smelters is estimated to be approximately 70% (Mangeng and Mead, p. 59). Through process modification or improved tail-gas treatment it is estimated that an average SO_2 control of 90% could be reasonably achieved at a cost of approximately $150 per ton of SO_2 removed (Work-Group 3B Report, pp. 117-137). This translates to a 66% reduction at $675/ton, relative to current emissions.

- Cement Industry

Through use of high-performance fabric filters on emissions from cement kilns, improved capture of SO_2 can be achieved via reaction with alkaline cement residuals (Work-Group 3B Report, p. A-17). Approximately 30% of existing kilns currently use fabric filters for particulate control. By upgrading the remainder, it is estimated that 5.1 lb SO_2/ton of cement produced could be reduced (AP-42) at a cost of $1.7-2.3 per ton of cement.

- Petroleum Refining

Two control options were included for refineries: switching from average sulfur-content fuel oil of 1.0% S to 0.3% S in refinery process heaters and improved 4-stage Claus Plants for high-performance tail-gas clean-up. Process changes to moving-bed fluid catalytic cracking were found to be highly cost-ineffective in reducing SO_2 emissions.

- Other Non-Utility Sectors

Sulfur recovery plants were assumed to achieve 99% reduction in sulfur emissions that would otherwise be flared in natural gas processing plants. These high-cost systems translate to a reduction effectiveness of >$7000 per ton of SO_2 removed. By shifting from single-contact control plants to double-contact plants in sulfuric-acid manufacturing facilities, SO_2 removal can be improved from about 98% to 99.7%. The cost of a typical plant is $9 million (1976 $), which translates to an 85% reduction over current emissions at a cost of $5940/ton (1979 $). The only feasible option for reduction of emissions in the iron and steel industry appears to be a process change to direct reduction which effectively eliminates all sulfur emissions. The cost of direct reduction systems is estimated to be $3.2/ton of steel produced, translating to >$10,000 per ton of SO_2 removed. Reduction of sulfur content of gasoline is considered to be the only major option for reduction of transportation-derived SO_2 emissions. About 70% of crude oil is considered amenable to treatment by sulfur absorbers prior to hydrocracking. About 80% of the sulfur can be removed by these systems. However, the costs of these absorbers were unavailable to us due to their proprietary nature. An estimate of $8000 per ton was used.

- Summary

The series of reduction potentials and costs are listed in Table B-5. The values for industrial boilers are considered quite reliable, even through averaged over a range of real-world cost and removal values. The smelter and cement industry options are also reliable because the control technology options considered have already penetrated the industrial sectors. The other control options are more speculative. Although based on engineering calculations of real control systems, they represent applications that are either not currently utilized or not demonstrated on large scale. More detailed engineering studies are needed to refine these numbers.

Because of uncertainty over the higher cost numbers in Table B-5, the $4400/ton cut-off was instituted, as discussed in the text.

APPENDIX C
USE OF ATMOSPHERIC TRANSPORT MATRICES

Several models have been formulated to relate sulfur dioxide emissions to wet and dry sulfur deposition rates. In the U.S.-Canadian Work Group 2 study, matrices relating 40 emission areas to 9 sensitive receptor areas were developed for seven different models using 1978 meteorological data. The models are: AES, ASTRAP, ENAMAP, MEP, MCARLO, OME, and UMACID. Table C-1 reproduces a sample matrix (for ASTRAP). Note the overprinted message advising caution in the use of these matrices. Table C-2 identifies the source regions for these matrices.

All of these models assume linearity between emissions and deposition (i.e., if emissions from a given source region were to drop 30%, the models predict that deposition in each receptor area due to that emission region contribution will drop an equal 30%). The National Research Council[7] has recently concluded that there is no convincing evidence of significant nonlinearity in eastern North American if averaged on an annual basis over spatial scales of about 1000 km or more.

For any given receptor area, the model results generally differ from one another by a factor of three or four when predicting total sulfur deposition (Figure C-1). However, as the main text shows, the models are more consistent in predicting changes in deposition (in percentage terms) relative to a baseline of total deposition.

For the purposes of this analysis, only six of the seven models were used, MEP being omitted on the recommendation of EPA. In comparing the predictions of the models with monitored wet deposition, it was generally observed that the AES and ASTRAP models came closest to reproducing current patterns of deposition. As the main text explains, the ASTRAP model was subsequently used with improved meteorology (1980 and 1981 annual average) as the basis for much of this analysis.

Table C-1 Sample Atmospheric Transport Matrix for the ASTRAP Model Using 1978 Meteorology

DRY DEPOSITION UNIT MATRIX IN KG.S/HA/TBS

REGION		BW	ALB	MUSK	QUEB	SNSC	VT	ADIR	PENN	SHOK
10	NMN	0.27	0.10	0.04	0.07	0.01	0.03	0.02	0.01	0.0
11	SMN	1.46	0.47	0.20	0.08	0.01	0.06	0.06	0.07	0.01
12	NWC	0.56	0.44	0.24	0.17	0.04	0.11	0.11	0.06	0.01
13	NEO	0.01	5.50	1.93	0.63	0.16	0.47	0.53	0.29	0.01
14	SUD	0.0	0.26	2.51	1.59	0.47	1.33	1.39	0.28	0.0
15	SWO	0.01	0.27	1.78	1.07	0.56	1.43	1.27	0.19	0.01
16	SEO	0.0	0.02	1.30	2.90	0.92	2.60	3.09	0.26	0.0
17	SLV	0.0	0.0	0.0	13.30	1.76	2.82	0.91	0.0	0.0
18	MOU	0.0	0.04	0.58	1.76	0.51	1.30	0.81	0.13	0.0
19	SBY	0.0	0.0	0.0	0.03	0.68	0.02	0.01	0.0	0.0
20	MBK	0.0	0.0	0.0	0.02	1.99	0.01	0.01	0.0	0.0
21	NSP	0.0	0.0	0.0	0.01	0.39	0.01	0.01	0.0	0.0
22	NFL	0.0	0.0	0.0	0.01	0.01	0.01	0.0	0.0	0.0
23	ASK	0.31	0.04	0.01	0.0	0.0	0.0	0.0	0.01	0.01
24	BCA	0.01	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
30	OHO	0.01	0.20	0.88	0.42	0.43	0.68	0.63	4.35	0.04
31	ILL	0.06	0.49	0.47	0.25	0.14	0.30	0.39	0.61	0.23
32	PEN	0.0	0.05	2.44	0.61	0.82	1.33	2.36	11.51	0.01
33	IND	0.02	0.34	0.71	0.27	0.28	0.43	0.61	1.49	0.15
34	KEN	0.0	0.11	0.34	0.15	0.23	0.30	0.46	1.47	0.44
35	MCH	0.03	0.38	0.54	0.45	0.31	0.65	0.82	0.76	0.01
36	TEN	0.0	0.02	0.11	0.05	0.11	0.12	0.21	0.79	5.61
37	MSU	0.11	0.56	0.50	0.19	0.07	0.20	0.28	0.40	0.21
38	WVR	0.0	0.04	0.18	0.24	0.40	0.46	0.45	2.85	0.03
39	NYK	0.0	0.01	0.16	1.01	1.35	4.51	7.37	0.37	0.0
40	ALA	0.0	0.02	0.05	0.02	0.03	0.04	0.08	0.34	3.68
41	WIO	0.37	1.45	0.73	0.25	0.09	0.24	0.29	0.36	0.07
42	MIN	1.94	0.71	0.36	0.13	0.03	0.11	0.14	0.17	0.03
43	VNC	0.0	0.02	0.14	0.16	0.38	0.29	0.37	1.28	0.16
44	FLO	0.0	0.01	0.02	0.01	0.02	0.02	0.04	0.14	0.29
45	GSC	0.0	0.01	0.05	0.04	0.12	0.08	0.12	0.28	4.21
46	MDJ	0.0	0.02	0.13	0.34	0.67	0.66	0.98	1.32	0.0
47	ALM	0.01	0.08	0.13	0.03	0.04	0.05	0.09	0.28	1.90
48	MCR	0.0	0.01	0.03	0.38	2.92	1.19	0.37	0.04	0.0
49	NAN	0.0	0.0	0.0	0.13	6.34	0.38	0.02	0.0	0.0
70	VNH	0.0	0.0	0.01	0.76	3.92	7.82	0.85	0.01	0.0
71	WNE	0.61	0.14	0.05	0.01	0.0	0.01	0.01	0.02	0.01
72	WSE	0.26	0.15	0.08	0.01	0.0	0.02	0.03	0.05	0.05
73	WNW	0.03	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
74	WSW	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0

THE OUTPUTS OF ALL ATMOSPHERIC SIMULATION MODELS ARE SUBJECT TO ERRORS AND UNCERTAINTIES OF VARYING DEGREE AND SIGNIFICANCE DEPENDING UPON THE VARIABLES BEING MODELLED AND THE SPECIFIC ANALYTICAL OR PREDICTIVE APPLICATIONS BEING CONSIDERED. TRANSFER MATRICES SHOULD NOT BE USED WITHOUT AN APPRECIATION OF THEIR LIMITATIONS AS OUTLINED IN THE ACCOMPANYING TEXT OF THIS REPORT.

Source: U.S.-Canadian Work Group 2 Draft Report

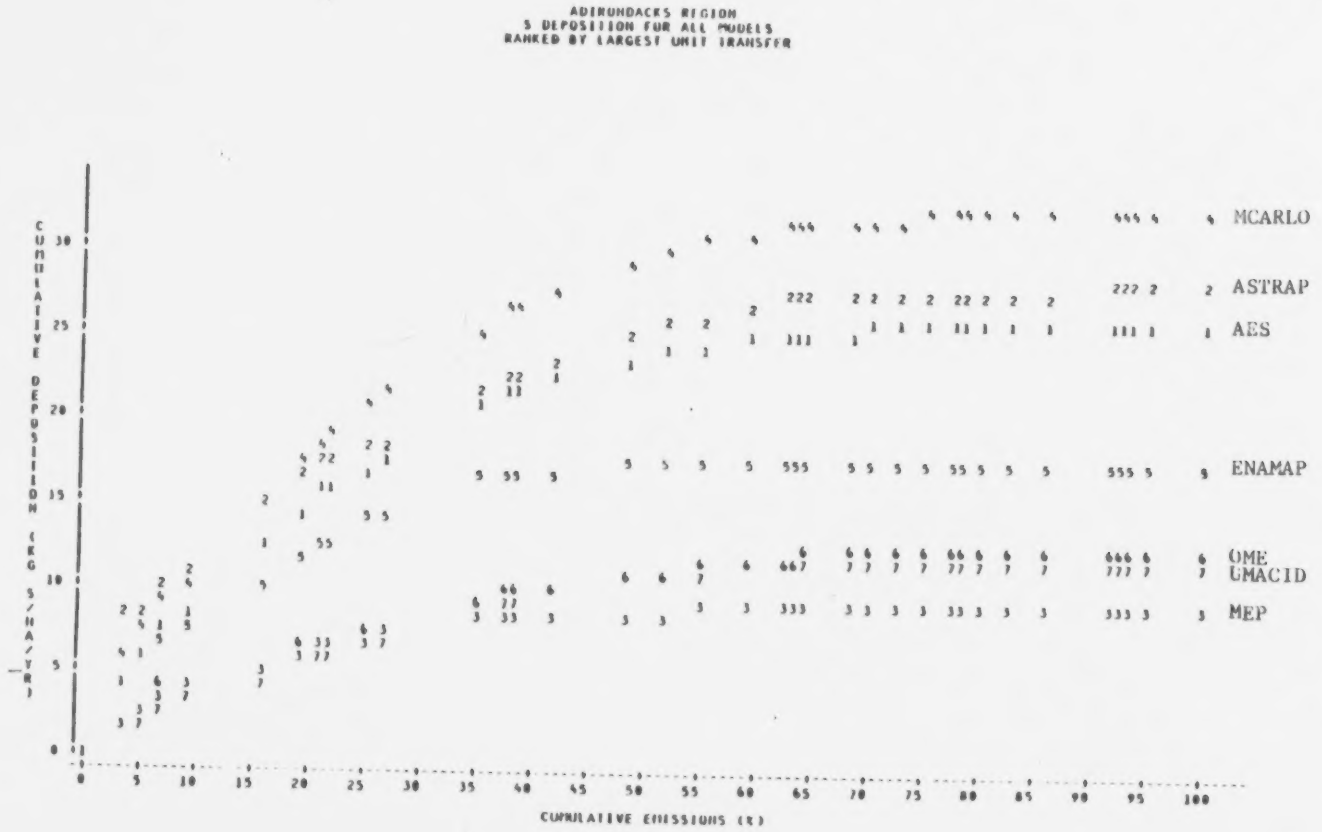
Table C-2 Identification of the WG2 Source Regions

Region Number	Emission Region	Region Centroid Latitude	Region Centroid Longitude
10	Northern Manitoba	56.84°	97.77°
11	Southern Manitoba	51.21°	98.34°
12	Northwestern Ontario	52.02°	89.20°
13	Northeastern Ontario and South Algoma	49.03°	83.03°
14	Sudbury	46.32°	80.70°
15	Southwestern Ontario and Toronto	43.56°	81.24°
16	Southeastern Ontario	45.23°	77.40°
17	Montreal and St. Lawrence Valley, Quebec	46.88°	72.13°
18	Noranda and North-Central Quebec	54.28°	73.59°
19	Gaspe Bay, Quebec	50.02°	64.56°
20	New Brunswick	46.21°	66.77°
21	Nova Scotia and Prince Edward Island	45.46°	63.27°
22	Newfoundland and Labrador	51.43°	58.89°
23	Saskatchewan and Alberta	50.0°	110.0°
24	British Columbia and Yukon/N.W.T.	49.3°	123.0°
50	Ohio	40.2°	82.5°
51	Illinois	40.0°	89.0°
52	Pennsylvania	41.0°	77.7°
53	Indiana	40.0°	86.1°
54	Kentucky	37.5°	85.5°
55	Michigan	44.5°	84.5°
56	Tennessee	35.9°	86.0°
57	Missouri	38.5°	92.5°
58	West Virginia	38.5°	80.5°
59	New York	42.7°	76.0°
60	Alabama	33.0°	86.8°
61	Wisconsin, Iowa	43.0°	92.0°
62	Minnesota	46.2°	94.5°
63	Virginia, North Carolina	36.5°	78.5°
64	Florida	29.0°	82.0°
65	Georgia, South Carolina	33.4°	82.8°
66	Maryland, Delaware, New Jersey, D.C.	39.2°	75.7°
67	Arkansas, Louisiana, Mississippi	33.2°	91.4°
68	Massachusetts, Connecticut, Rhode Island	42.1°	72.2°
69	Maine	45.5°	69.0°
70	Vermont, New Hampshire	43.7°	72.5°
71	Central Northwest States (1)	45.0°	104.0°
72	Central Southwest States (2)	34.2°	100.0°
73	Pacific Northwest States (3)	45.0°	118.4°
74	Pacific Southwest States (4)	37.3°	115.7°

(1) includes: Nebraska, North and South Dakota, Montana, Wyoming.
(2) includes: Oklahoma, Kansas, Colorado, New Mexico, Texas.
(3) includes: Washington, Idaho, Oregon.
(4) includes: California, Nevada, Utah, Arizona.

Source: U.S.-Canadian Work Group 2 Draft Report

Fig. C-1 Comparison of Sulfur Deposition Models



APPENDIX D
SENSITIVE RECEPTOR REGIONS

D.1 Geographical Locations

Nine sites were used in the U.S.-Canadian study to identify locations in northeastern North America which are sensitive to acidic deposition. These are listed in Table D.1. Figure D.1 shows these locations on a map of North America. These are the target regions considered in this analysis.

TABLE D.1 RECEPTOR COORDINATES* FOR THE TARGETED SENSITIVE AREAS

Area Number	Name	Latitude	Longitude
1	Boundary Waters	49.0°	93.0°
2	Algoma	46.5°	84.0°
3	Muskoka	45.0°	79.5°
4	Quebec City	47.0°	72.0°
5	Southern Nova Scotia	44.0°	66.0°
6	Vermont/New Hampshire	45.0°	72.0°
7	Adirondacks	44.0°	74.0°
8	Western Pennsylvania	41.0°	78.0°
9	Southern Appalachia	35.0°	84.0°

*The receptor sites may vary by $\pm$ 0.5° due to grid resolution of the respective models.

Source: U.S.-Canadian M.O.I. Study

D.2 Target Loadings

Target loadings are numerical goals for an acceptable or "safe" rate of acid deposition. The difficulty with target loadings, of course, is selecting an appropriate number (or range for specific conditions). To date, target loading recommendations have generally been set with respect to protection of aquatic resources. One exception is the June 1982 Stockholm Conference report, which suggested tree growth may be affected by annual SO_2 concentrations of 25-50 $\mu g/m^3$, with episodes exceeding 100-200 $\mu g/m^3$. Since some lakes have been acidified due to deposition loads of the recent past, while crop and forest effects are only at the hypothetical stage, it would appear reasonable to assume that deposition loadings acceptable for lakes will generally protect forests and crops. This discussion will continue the trend of focusing on loading targets for protection of aquatic resources.

As part of the work conducted pursuant to the U.S.-Canadian MOI, Canadian members of Work Group I proposed a deposition rate of 20 kg SO_4/ha-yr (in precipitation) as protective of all but the most sensitive lakes. "Most sensitive" was postulated as lakes with alkalinity less than 40 $\mu eq/l$. US members of the Work Group concluded, to the contrary, that no target loading for sulfur or nitrogen compound deposition could be derived from current information and that for the specific areas having data, 20 kg SO_4/ha-yr was too low by at least 10 kg.

Dickson (Swedish Environmental Protection Board) used an empirical model to predict a "safe" loading of 0.3 g S/m^2-yr (9 kg SO_4/ha-yr) (bulk) to protect the most sensitive Swedish lakes, although he noted that 3-5 times that rate appeared acceptable for less sensitive Swedish lakes. He did not define sensitive as a value but instead the lower volume of ANC (alkalinity) found in each region. (These varied by 400% from region to region).

The U.S. National Academy of Sciences recommended a precipitation pH of 4.6-4.7 as "desirable" (Atmosphere-Biosphere ___, 1981). Hence, this "target" explicitly incorporates acid species other than sulfur. This pH target obviously includes only wet deposition.

No group has proposed acceptable rates for dry deposition of acidic compounds, but most investigators conclude such deposition is as important as a chemically equivalent amount of wet deposition.

To make sense of the different target objectives, they must be converted to equivalent terms. These conversions are presented in Table D-2. Data for Woods Lake, in the Adirondacks, were used to make the conversions (see Characterization of Acid Precipitation in the Adirondack Region, EPRI, 5/81). (Woods Lake is presently acidified, e.g., ANC less than zero).

Table D-2 demonstrates that "goals" published to date differ by about a factor of 5. This difference probably results from subjective decisions regarding the impracticality of protecting extremely sensitive lakes.

The influence of natural sulfur emissions has been ignored in this analysis. It is believed to be small (causing 1-10% of total sulfur deposition in the Adirondacks). However, if natural sulfur deposition is as high as 10%, this implies that the "targets" will be significantly more difficult to attain, and the 91% reduction suggested by Dickson would be unattainable.

It is important to consider the basic assumptions underlying target loadings if they are to be applied prudently. The targets are for annual deposition, and they are designed to protect aquatic resources. However, fish population stress, caused primarily by interference in reproduction or fry kills, are observed almost entirely in the spring when annually averaged characteristics may not be valid. Acid loadings to lakes surge as winter snows melt. An additional consideration is that nitrogen-associated acidity

may equal or dominate sulfur acidity in the spring surge period, whereas sulfur dominates over an annual period. For these reasons annual targets, especially targets based only on sulfur deposition, could be unreliable as guides to protect sensitive resources.

Two factors limit general application of the conversions provided in Table D-2.

- The first is the relationship between sulfate and total hydronium ion concentration in water. The two parameters do not correlate simply over large numbers of lake samples. This precludes area-based sulfate to pH conversions for use of sulfur deposition as a valid acidic deposition surrogate for environmental management purposes.

- The other problem is the ratio of wet to dry deposition. Dry deposition is not accurately measured, but different models examining different geographic areas indicate a potentially large variation in wet/dry ratios (from 10:1 to less than 1:1). This affects conversions of total deposition to "wet only" ratio or pH concentrations.

This study examines two of the proposed target loadings: 20 and 30 kg wet sulfate Ha^{-1} yr^{-1}. This method is clearly applicable to any target concept of this kind.

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Table D-2 Deposition Reduction Targets Proposed in Various, Selected Reports

Source	Target As Published	Target Converted To: gS/m2-yr (total)	kg SO4/ha-yr (wet)	pH*	Reduction in S Deposition Needed to Attain Target (%)
1. NAS	4.6-4.7 (pH)	0.8-1.1	9.5-12.5	4.6-4.7	68-75
2. CAN-WG-1	20 kg SO4/ha-yr	1.7	20.	4.4	48
3. Dickson (most sensitive)	0.3 gS/m2-yr(total)	0.3	3.6	5.0	91
(less sensitive)	0.9-1.5 gS/m2-yr (total)	.9-1.5	11-18	4.5-4.7	54-72

*Based on Woods Lake chemistry June 1978 - May 1979 (Adirondacks)

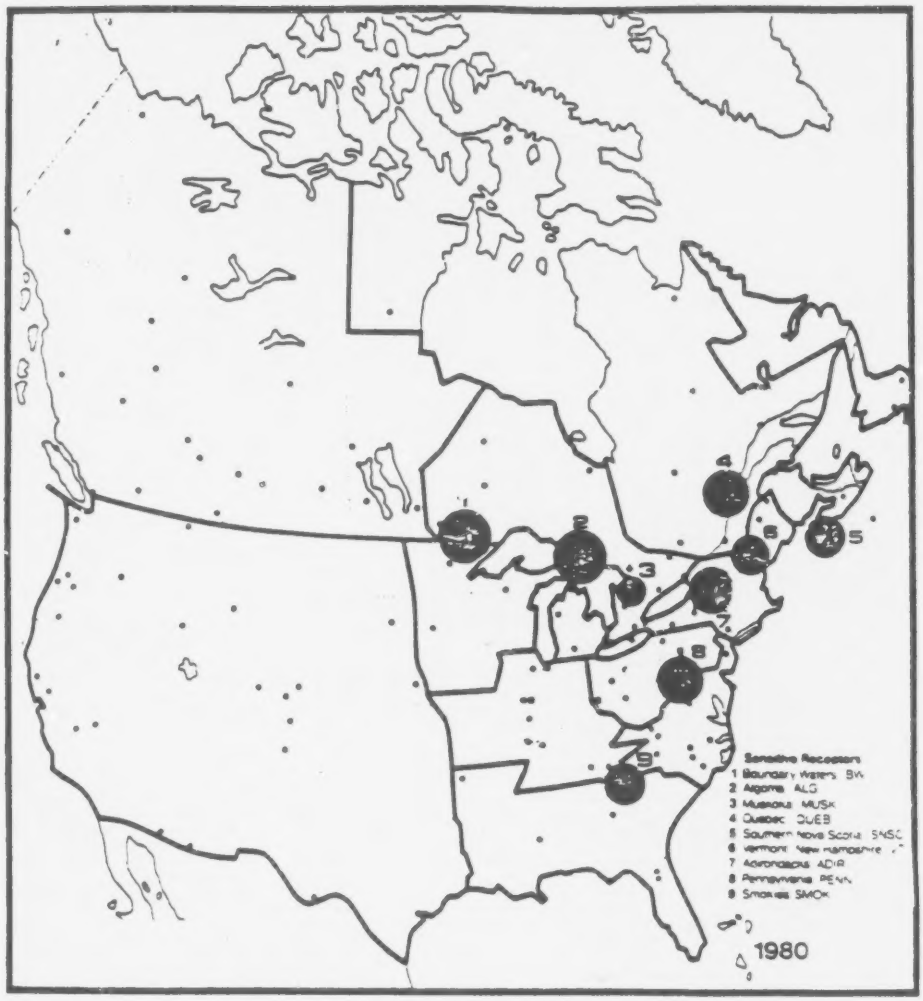

Fig. D-1 Locations of Sensitive Receptor Areas

DON FUQUA (Fla.), Chairman

ROBERT A. ROE, N.J.
[illegible]
JAMES H. SCHEUER, N.Y.
[illegible]
MARILYN LLOYD, TENN.
DOUG WALGREN, PA.
DAN GLICKMAN, KANS.
ALBERT GORE, JR., TENN.
[illegible]
HAROLD L. VOLKMER, MO.
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HALEY M. HALL, TEX.
[illegible]
MIKE LOWRY, WASH.
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PAUL MCHALE, PA.
RICHARD J. DURBIN, ILL.
MICHAEL A. ANDREWS, TEX.
BUDDY MACKAY, FLA.
TIM VALENTINE, N.C.
HARRY M. REID, NEV.
ROBERT G. TORRICELLI, N.J.
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BROOKHAVEN NATIONAL LABORATORY
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Department of Applied Science

(516) 282- 3037
FTS 666-

August 30, 1983

The Honorable Don Fuqua
The Honorable James H. Scheuer
The Honorable Marilyn Lloyd
U.S. House of Representatives
Committee on Science & Technology
Suite 2321, Rayburn House Office Bldg.
Washington, DC 20515

Dear Representatives:

In response to your letter of August 4, 1983, I have asked the National Laboratory Consortium to express their views on the results stated in the National Academy of Sciences report entitled "Acid Deposition-Atmospheric Processes in Eastern North America" which was authored by an NRC Committee.

The four National Laboratories which make up the National Laboratory Consortium are Argonne National Laboratory, Brookhaven National Laboratory, Oak Ridge National Laboratory, and Pacific Northwest Laboratory of Battelle. The attached documents from each of the National Laboratories provide critiques of the National Academy of Sciences report by individual scientists at the National Laboratories each of whom is identified. Please note that, in each case, these comments have been reviewed internally by appropriate Laboratory authority. In sum, they therefore represent the collected opinions of the National Laboratory Consortium.

As one reviewer states, "the Committee has made a useful contribution by bringing before the public ...in rather the most effective fashion to date... findings of many years of research on the transport, transformation, and deposition of airborne pollutants." Although there are many specific instances in which the Committee has brought together disparate sources of information and reached logical conclusions, the overall feeling of the National Laboratory Consortium is that the Committee's report is flawed in several areas.

A principal conclusion of the NRC Committee regards the relation between emissions of sulfur dioxide and deposition. The chairman of the NRC states in his forwarding letter that it is the "Committee's judgment that if the emissions of sulfur dioxide from all sources in this region (northeastern North America) were reduced by the same fraction, the result would be a corresponding fractional reduction in deposition." The National Laboratory Consortium reviewers disagreed almost uniformly with the methods used to reach this conclusion and with the conclusion itself.

Specific and detailed comments on the NRC documents are contained in the reviews written by National Laboratory scientists, all of whom are active in the field.

Major points made by the reviewers are directed at two technical areas. The first relates to the interpretation of the spatial dependence of the sulfate-to-nitrate ratio.

- Data presented on sulfate-to-nitrate ratios in precipitation and emissions do show spatial patterns in the same direction and of the same magnitude that are used to justify a nonlinear relationship in Europe. Even accepting a constant ratio, the conclusion of a linear removal mechanism for sulfur dioxide is not supportable. (Reviews BNL-A, PNL)

- Details of gas phase chemical reactions and in particular the deficiencies of Rodhe's photochemical model are given more weight that can be justified in regard to a chemical-mechanistic interpretation of linearity in the atmosphere. Nonlinearities are more likely to result from pH inhibition in aqueous phase reactions or from oxidant limitations; topics not adequately covered. (Reviews ANL, BNL-B, BNL-C, BNL-D, PNL)

- The report concerns itself almost entirely with large spatial areas (Federal Region to sub-continent) and long averaging periods (seasonal to yearly). Averaging over such time and space scales obscures a wealth of detail that is pertinent to the mechanisms of acid deposition. A closer look at finer spatial scales is particularly important in regard to any discussion of linearity. If large enough spatial scales are chosen, conservation of mass ("what goes up must come down") insures that deposition is linearly related and, in fact, equal to emissions. Furthermore, the million-square-kilometer mixing that is discussed occurs only as an artifact of averaging over long time periods. (Reviews ANL, BNL-A, BNL-B, BNL-D)

The second technical area relates to the use of the long term record at Hubbard Brook, NH:

- o The Academy report attempts to relate the deposition trends at Hubbard Brook to Northeastern emission trends in a very superficial way, without confidence limits on the findings. Detailed statistical reanalysis of Hubbard Brook data, using multiple regression analysis to account for meteorological variability over the period of record as well as emission estimates for every year, finds a substantially less than 1:1 proportionality between sulfate deposition and sulfur oxide emissions. This lack of proportionality suggests a nonlinear relationship. (Reviews ANL, BNL-A, BNL-C, ORNL-A, PNL)

- o The historical records of wet deposition at Hubbard Brook, NH, in relation to emissions trends in the Northeast, plays a major role in the arguments for "linearity" (and implied proportionality). Since Hubbard Brook is well outside the region of maximum emission and deposition, it is unlikely that experimental data could reveal whether processes are linear or not. (Reviews BNL-B, PNL)

- o A significant proportion of the sources that affected Hubbard Brook were probably located in southeastern Canada; yet Canadian emissions were not included in the report's trend assessment. (Reviews ANL, BNL-A)

And finally:

- o A lack of evidence for nonlinearity in the empirical data examined by the Committee is not logically equivalent to a proof of linearity. (Reviews BNL-C, PNL)

On the other hand, the National Laboratory Consortium is in substantial agreement with the Committee's stated research priorities and needs. Directed field experiments, both mechanistic and data gathering, are suggested to have a first priority. The report further explains that this priority is based upon the expectation that such experiments would yield results in a shorter time than required for a molecular and dynamic description; however, laboratory and model results are needed and these activities should also proceed. Other suggestions with which we concur are the determination of source-receptor relations via atmospheric tracer and computer air parcel trajectories and the need to develop a routine monitoring method for dry deposition.

We believe that most of the Committee's assessment of research needs are incorporated in the program of the Interagency Task Force on Acid Precipitation. If that program is fully implemented, many of the uncertainties related to the most significant issues should be substantially reduced or resolved.

Sincerely,

Bernard Manowitz
Chairman, National Laboratory
Consortium of the Interagency Task
Force on Acid Precipitation

BM:mh
atts. 10
cc: Frank Press, Chairman, NRC
Herbert Friedman, Chairman, Comm. on Phys. Sci., Math & Res.
Jack Calvert, Chairman, NRC Comm.

Review ANL

ARGONNE NATIONAL LABORATORY
9700 South Cass Avenue, Argonne, Illinois 60439

Telephone 312/972-

July 26, 1983

Mr. Bernie Manowitz, Chairman
National Laboratory Consortium
Bldg. 179A, Department of Applied Science
Brookhaven National Laboratory

Dear Bernie:

The following summarizes our Atmospheric Physics Section's comments on the report entitled "Acid Deposition: Atmospheric Processes in Eastern North America", prepared by the Committee on Atmospheric Transport and Chemical Transformation for the National Research Council. Because of his broad involvement in many aspects of the National Acid Precipitation Assessment Program, most of this commentary is due to Dr. Jack Shannon, Leader of the Section's Modeling Group. Discussions have also included Dr. Marvin Wesely, Dr. In Young Lee, Dr. Romesh Kumar, and myself.

By and large, the Committee has made a useful contribution by bringing before the public--in rather the most effective fashion to date--findings of many years of research on the transport, transformation, and deposition of airborne pollutants. Nevertheless, we do have some differences of opinion relative to the Committee's conclusions, differences that are primarily matters of scale and degree. But at this stage of developing knowledge of these complex atmospheric processes, and at this point in the developing consensus for some as yet poorly defined legislative action, these differences could have far reaching consequences.

The NRC/NAS report makes a number of conclusions and recommendations which we believe are either not convincingly supported by the evidence presented, or for which we prefer alternative views supported by the evidence of our own work. These include statements such as:

1) On an annual time scale and over a 1000-km spatial scale, the formation and deposition of sulfate do not appear to be significantly nonlinear.
2) Current operational models of transport and deposition are inadequate for policy analysis.
3) Relative contributions of specific source regions to specific receptor sites are unknown.

Since each of these points deals in some way with the question of the relative "validity" of current models, it is difficult to discuss them separately. The following attempt to do so proceeds in the order presented above.

The NRC/NAS report limits its finding of "no significant nonlinearity" to the larger scales given in 1), above. Our reservations in this regard are concerned with the improved definition of these scales, specifically the determination of the shortest transport times and smallest distances below

US Department of Energy The University of Chicago Argonne Universities Association

which neglecting nonlinearity could have important consequences. Time scales over which nonlinearities may affect policy should be accomodated in the design of emission control strategies. Clearly these scales are related to ecological and material effects; effects on lakes and watersheds do seem to be largely long-term--perhaps seasonal, as in snowmelt shock. But if shorter term ecological "insults" are found to be important in current research (for example, effects on vegetation at critical stages of growth), then the magnitude of time scales for nonlinearity would need reexamination.

The 1000 km spatial scale is much larger than the distances between many combinations of source and sensitive-receptor areas. The possibility of nonlinearity over shorter distances would therefore have to be considered for the consequences of potential emission control policies to be assessed accurately. Then too, even if a uniform rollback policy were adopted, all emission sources could not be uniformly reduced because of economic or equipment constraints. Instead, rollback amounts would probably be defined over some political entity of medium scale, such as a state. Current emission levels per BTU vary widely now, and some sources are much more amenable to emission control actions than others. For instance, if a plant has already installed a scrubber, little additional emission reduction could be required of that plant if it is to continue to operate. Thus, rollback could not be expected to be uniform even when totaled by state. In short, the NRC/NAS report appears to address the issue of deposition reduction by control legislation too narrowly.

We agree with the report's conclusion that nonlinearities are more likely to occur in winter, when many transformation and removal rates are slower and transport generally faster. But even this is a qualified agreement pointing up the issue of scale: specifically, over just what distances would nonlinearity be expected to limit the efficacy of rollback legislation in these--or any--weather conditions? The consequences of the interpretation of nonlinearity are too far-reaching to be based on the limited data used.

The evidence cited by the NRC/NAS report in support of their argument for linearity is a) the ~ 15-year record of weekly bulk deposition collected at Hubbard Brook, New Hampshire, b) historical SO_2/NO_x emission estimates , and c) the observed spatial patterns of molar ratios of SO_2/NO_x emissions and sulfate/nitrate wet deposition. An apparent trend of decreasing sulfate deposition at Hubbard Brook is associated with a trend of decreasing SO_2 emission in the Northeastern U.S. But there are several difficulties with this analysis:

1. No uncertainty ranges are placed on any of the emission totals. Since the SURE, MAP3S, and MOI inventories for a single year (1978) show differences in total emissions by states of 30 to 40 percent, the historical emission data used may be equally--or even more--uncertain. Such uncertainties would swamp the apparent effects of any actual emission change.

2. Trends in Canadian emissions are not considered at all; yet the Canadian contribution to bulk deposition received in central New Hampshire, i.e., at Hubbard Brook, is probably about 25 to 40 percent of the total.

3. The apparent decrease in sulfate deposition at Hubbard Brook largely results from reference to relatively high values recorded in the initial years

(approx. 1965-1969). Since that time, there appears to have been little or no further decrease in sulfate deposition, even though emission inventories over subsequent years did continue to decrease. Meteorological variability could have accounted for this deposition record, even if there had been no change in emissions; or a single, not-too-distant source operating only in the earlier years may have gone unnoticed.

4. The decrease in the molar ratio of the sulfate/nitrate bulk deposition at Hubbard Brook also seems to result primarily from a high ratio recorded in the late 1960s followed by irregularly lower values in subsequent years. The Hubbard Brook data suggests a step function rather than a trend. The possibility of analytical error--or a change in analytical technique--could be considered.

5. The NRC/NAS report does not resolve the paradox of the apparent evidence for linearity in the Hubbard Brook data and the evidence for nonlinearity in the European data. The comparison between statewide SO_2/NO_x molar emission ratios and sulfate/nitrate wet deposition ratios used to support the linear hypothesis, is not conclusive. For example, some preliminary results of modeling sensitivity tests now underway at this Laboratory suggest that the sulfate/nitrate wet deposition molar ratio is relatively insensitive to the SO_2/NO_x emission ratio; thus a decrease in SO_x emissions may not be proportionally reflected in a subsequent decrease in sulfate wet deposition.

6. The discussion concerning whether or not the Rodhe model is adequate without a chain-carrying radical to continue the reaction, glosses over the limitation imposed by the fact that both versions of the set of equations used are essentially clear-air chemistry models, and are therefore not directly relevant to wet deposition processes.

With regard to present models, we would suggest that the relative utility of many recent assessments indicates that a number of the numerical simulations now in use have been demonstrated to be adequate for policy analysis. Future improvements in modeling now proposed (and evidently advocated for future use in assessment by the NRC/NAS report) are more likely to improve diagnostic details and field experiment design than contribute directly to improvements in policy assessment exercises. Yet the report recommends against the use of current transport and deposition models for policy assessment, thus recommending , in effect, that no cost/benefit assessment be done at this critical time of legislative decision. These negative views on present modeling capability seem to be based upon the level of parameterization necessary for tractable models, and upon the Committee's perception of the results of the now outdated MOI model intercomparison.

Taking the second point first, there were many uncertainties in the 1978 SO_x emission inventory for North America used by the MOI comparison. Further, the precipitation chemistry samples used for model verification were monthly collections and were almost entirely derived from southeastern Canada. Finally, most of the models compared had only two months of 1978 meteorological data available to them. It follows that the MOI model intercomparison was hardly a rigorous test, especially in view of the improved data sets (not to mention the improved models) available today. The NRC/NAS report does concede that some present models "quantitatively point to the importance of certain geographical groupings of SO_2 sources and the potential

influence of (those) sources on certain receptor areas". It can be added that some of today's parameterized models consistently reproduce, in realistic fashion, average (monthly and longer) downstream deposition patterns attributable to large groups of sources, in the form of so-called source-receptor matrices. The "ASTRAP" model developed and used at this Laboratory, for example, achieves this degree of success with objective parameterizations of mixing, transformation, and removal processes which are based on actual measurements made in ad hoc field experiments. In other words, the model has not been arbitrarily tuned to attain this performance.

This brings us to the third point of difference. On the basis of our modeling experience, we believe that average monthly and seasonal contributions of specific source regions of significant size (say, half the area of a medium sized state) to receptor regions of similar size up to 1000-1500 km downstream, can be simulated with present models sufficiently well to make objective assessments. But until the emissions from particular source regions can be identified in deposition samples, or until large scale field experiments can be performed with realistically reacting/depositing tracers, no transport/deposition model of source/receptor relationships can be rigorously verified. Present models use real fields of meteorological data, after the fact. Indeed, it is recognized that whenever possible, assessments over a certain period (month, season) should be based on models run with several years of data in order to limit the effects of meteorological anomalies. We do not believe that there is any realistic need for a truly dynamic transport and deposition model that would operate in a meteorologically predictive mode.

Even with some uncertainties in detail, analyses of proposed emission control policies based on current models are likely to be far superior to analyses of control policies without such objective consideration of the complex source/receptor relationships. Since some emission control actions may well be taken soon, it is critically important that current knowledge of transport and deposition processes, albeit imperfectly represented in today's models, be utilized in the best interests of both the public and the industry. Recommendation against the use--and, by implication, against the continued improvement of--the parameterized models, is therefore not in the national interest, particularly since the complex, 3-dimensional Eulerian grid models promised as an alternative will not be ready to simulate even a single, specific synoptic event for 2 or 3 years. Whether these complex computerized constructs will ever be sufficiently flexible--and sufficiently economical--to operate in the multiple-scenario, quick turn-around mode required for policy assesment remains to be seen. Even then, this will not be known until 3 to 5 years, hence when the new model attains a capability of simulating a statistically significant number of events.

Sincerely,

Paul Frenzen
Associate Division Director
Environmental Research Division

PF/lac
cc: J. Rundo, Associate Laboratory Director, OTD/BER
J. Roberts, Associate Laboratory Director, OTD/ELT

BROOKHAVEN NATIONAL LABORATORY

MEMORANDUM

DATE: August 29, 1983

TO: N. Samios, Director, BNL

FROM: B. Manowitz BM

SUBJECT: Review of National Academy of Sciences Document

In my capacity of Chairman of the National Laboratory Consortium of the Interagency Task Force on Acid Precipitation, I have been asked to review the document recently issued by the National Academy of Sciences entitled "Acid Deposition - Atmospheric Processes in Eastern North America."

I have asked four scientists in my Department to review the report. They are:

Dr. Fred Lipfert (identified as Reviewer BNL-A)
Dr. Paul Michael (identified as Reviewer BNL-B)
Dr. Lawrence I. Kleinman (identified as Reviewer BNL-C)
Dr. Stephen E. Schwartz (identified as Reviewer BNL-D)

These men were selected because of their long experience and expertise in air quality research (A), meteorological and atmospheric physics research (B), air chemistry modeling (C), and atmospheric chemistry research (D).

Their comments are substantive and critical of the broad conclusions of a 1:1 proportionality between emissions and deposition implied in the Academy report.

I concur entirely with the findings of these scientists.

pt

Review BNL-A

CRITIQUE OF CHAPTER 4 — EMPIRICAL OBSERVATIONS AND SOURCE-RECEPTOR RELATIONSHIPS

Introduction

Part of the thrust of the NAS report is that, given the present state of source-receptor modelling, we must rely on empirical observations to infer relationships between emissions and deposition and then to assess responses to mitigating strategies. One of the data sets presented for this purpose was the 16-year record of weekly wet deposition at the Hubbard Brook Experimental Forest in Central New Hampshire. While the emphasis on experimental observations may be appropriate at this time, the analysis presented was superficial and inappropriate to not only the Academy but to the importance and economics of the issue at hand.

The Hubbard Brook data have been extensively discussed and analyzed elsewhere.[1,2] But a serious attempt at a time series analysis relating these measurements to historical emissions of acid-forming precursors has not been presented, nor have quantitative relationships between wet deposition and air quality been assessed. Such a study is in progress and preliminary results are presented below.

The Academy Analysis (pp. 116-126)

The Academy analysis is based on a loose comparison of time trends between 1965-66 and 1979-80* for sulfate concentratons and emissions trends between 1965 and 1978 for any of several geographic regions. The emissions data are from the U.S. Canadian Work Group 3-B report,[3] which also reports data for two intermediate years, 1970 and 1975 (unfortunately, 1975 is a recession year and does not provide a reliable basis for trends between 1970 and 1978). Concentrations of nitrate in rain vis-a-vis NO_x emissions are also discussed, but more briefly. No statistical relationships are attempted between rain chemistry and emissions, nor are any confidence limits given for possible associations.

The trend in sulfate concentration (but not nitrate) is deduced from a linear regression analysis on 15 annual volume-weighted means, presumably giving each year equal weight. No explanation is given for excluding the two end periods, 1964-65 and 1979-80 (including them reduces the slope by about 10%). However, the trend in emissions over an equivalent period is deduced from only the two end points of the series, 1965 and 1978. The detailed behavior over the included time period is totally ignored as if the trend were monotonic (and linear) between these end points. Figure 4.15 shows that is decidedly not the case, even using the unrepresentatively low year of 1975 as a valid trend point. The fact is that 1970 (or thereabouts) is generally the peak year for SO_x emissions, after which the impact of the Clean Air Act of 1970 began to be felt. However, these reductions were realized slowly, so

*Hubbard Brook publications use a "water-year," beginning in June.

that the years 1968-1974 are practically flat for national SO_2 emissions. The decrease began sooner in the Northeast, about 1972. In contrast, Figure 4.14 shows an increase in $SO_4^=$ concentrations during this period. To be fair, however, it should be mentioned that a linear regression of the individual weekly samples shows an overall drop of about 20% over 15 years (discussed more extensively below).

In addition to using disparate analytical methods for the concentration trends vs the emissions trends, the Academy report does not deal with the question of estimating which geographic regions' emissions have the most direct impact at Hubbard Brook. By implication, the analysis points towards the nearby locations (Federal Regions I and II) and would totally exonerate Region IV, where emissions increased 33% from 1965 to 1978.

A similar process is followed for nitrate vis-a-vis NO_x emissions. Including the two end periods in a linear regression on time increases the change from +8% to +23% (increase with time). The emissions increase, calculated from only the two end points 1965 and 1978, was 8.6% for Federal Regions I and II, whereas the NO_x emissions increase for Regions I-V was 27%. Thus, the implication as to whether NO_x has an influence from areas further away than New Jersey is critically dependent on which measurement years are used at Hubbard Brook. Using all the data to determine the trend would imply that more distant source regions should be considered for NO_x (but not for SO_2), an argument that runs contrary to the discussion on sulfate/nitrate ratios in the Academy report.

The Academy report concludes, with regard to the Hubbard Brook data, that there is "no evidence for a strongly nonlinear relationship between annual depositions and annual emissions in the Northeast." As a result, elsewhere in the report, the finding is reached that a reduction in emissions would result in a proportional reduction in deposition. This implies a 1:1 relationship or an elasticity of 1.0 [a 1% change in emissions would yield a 1% change in deposition (or concentration)]. It should be clear from the data in the Academy report that this finding is not supported, even with the limited analysis presented.

For sulfate concentrations, the following elasticities result from the Academy analysis, depending on the region of SO_2 emissions to be controlled:

Region I	1.15
Regions I and II	1.18
Regions I and II and III	0.67
Regions I - V	0.25

Thus, given the uncertainties of the analysis, the linear (i.e., proportional) hypothesis is supported if only the emissions from Regions I and II are considered for control, but not if Regions further south and west are included. The bulk of the report (and most of the extant literature on source-receptor relationships) makes it clear that such more distant regions should in fact be considered.

To illustrate the relationship between elasticities (as deduced from linear regressions) and hypothetical non-linear relationships, consider Figure 1:

Figure 1(a) shows three linear relationships with differing elasticities. The proportional relationship, $\epsilon = 1.0$, yields a 50% reduction in concentration (or deposition) for a 50% change in emissions. However, if $\epsilon = 0.5$, only half the benefit is realized due to the presence of a y-intercept which could represent deposition arising from natural background or from sources outside the region of consideration. Converse arguments hold for $\epsilon = 2.0$.

Figure 1(b) shows how an actual non-linear relationship ($C = \sqrt{E}$) might be represented by linear approximation, for example derived from regression analysis. A linear regression through the entire curve (11 points spaced every 10% in emissions) yields a correlation coefficient of about 0.96, explaining 92% of the variance (which would be considered an excellent fit). The slope (and hence elasticity of this line is 0.87, which is not very different from unity. Yet if reductions in emissions are postulated of the order of 50%, the asymptotic linear approximation shows that only about half of the expected benefit would be realized in fact. It should be noted that in most cases, we are working with the upper ends of the curves in question (the range in National SO_x emissions from 1965 to 1979 is about 5 million tons out of a total of about 30 million.

It should be clear from this example that it is extremely unlikely that statistical analysis of time trends will be able to detect nonlinearities per se. However, differences in the elasticities of linear best-fit relationships should indicate the degree of proportionality to be expected. The range of elasticities given above, based on the figures in the Academy report, indicate that no conclusions on linearity or proportionality can be made from this analysis.

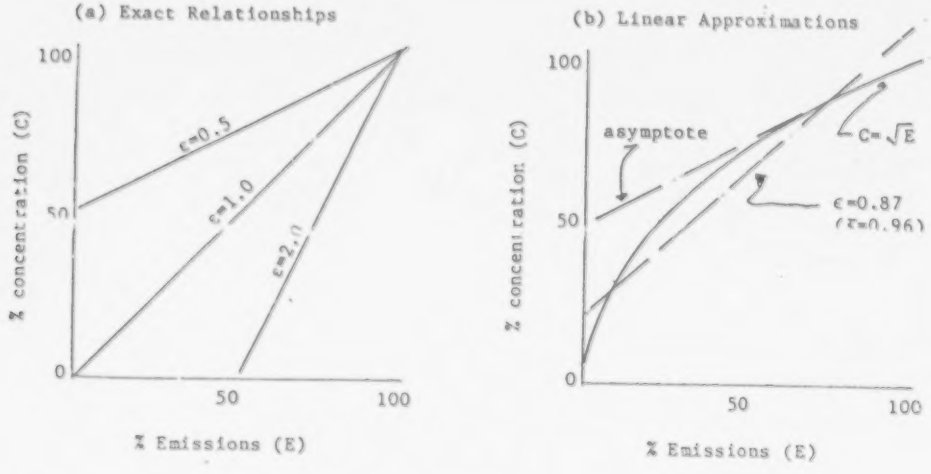

Figure 1

Reanalysis of the Hubbard Brook Precipitation Chemistry Data (1964-79)

Because of the importance of this data set, a reanalysis of the weekly (bulk) samples has been carried out, using multiple linear regression methods. The dependent variables were the amounts of precipitation in each weekly sample and concentrations (μeq/ℓ) of the following ions: H^+, $SO_4^=$, NO_3^-, Cl^-, Al^{+++}, NH_4^+, Ca^{++}, Mg^{++}, K^+, SiO_2, $PO_4^=$ and Na^+. Concentrations were corrected for sea salt content assuming that all the sodium is derived from that source. Further, the balance between positive and negative ions was calculated, and only the observations for which this balance was achieved within ±20% were retained (about 87% of the observations or about 440 values).

The following independent meteorological variables were used:

PRECIP — the amount of each weekly sample

WAVE — a periodic function intended to be a surrogate for such seasonal effects as photoperiod, wind trajectory variations, fraction of convective vs continuous storm, etc.

$$\text{WAVE} = \cos\left[\left(\frac{\text{day of year}}{365} \times 360\right) - 180 \ * \frac{\pi}{180}\right]$$

MAVTEMP — monthly average surface temperature at Concord, NH (finer time resolution not readily available).

The influence of trajectory differences by year at Hubbard Brook was considered in detail by Munn et al.[2] SO_4 concentrations were normalized by accounting for the frequency of occurrence of 5 different wind direction sectors; concentrations tended to be higher for trajectories originating to the south and west of Hubbard Brook than from the east, for example. However, this normalization had only a trivial effect on the actual annual median SO_4 concentrations, increasing the overall mean about 1-3%, but with virtually no change in the standard deviation from year-to-year. Thus, by implication, the differences shown from year-to-year in median sulfate concentration are not due to trajectory variations. The assumption that this source of variation may be ignored in a multiple regression analysis without creating bias appears to be well-founded.

The emissions data were computed for each year from 1964 to 1979 based primarily on fuel use data[4] at the state level for the United States. Statewide fuel sulfur contents were derived from FPC Form 67 data from 1969 to 1976. For years prior to 1969, these values were assumed constant. For years after 1976, emissions by state were estimated from published EPA reports. In general, this approach captures only combustion-derived emissions, a limitation that is not believed to be serious for the Eastern United States. Canadian copper-nickel smelter emissions[5] were also included for those years, as was an estimate of other eastern Canadian emissions,[6] for which only 2 years' data were available requiring linear interpolation to represent the 16 year period.

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Quarterly shares of emissions were available for 2 years (1965 and 1978),[9] for the SURE region (Northeastern U.S.). The differences between these shares and a uniform annual distribution were minor, with higher emissions in the winter. When used, these quarterly shares were applied to all emissions estimates, regardless of geographic region. A conservative estimate of the number of degrees of freedom for the emissions data is then 18 (16 annual values plus 2 quarterly shares). Comparison of these independent emissions estimates with those of EPA shows somewhat higher estimates but similar trends (for the years available from EPA).

As an alternative to aggregation by summation, weighting schemes were considered in order to combine emissions from the various geographic regions thought to have an influence at Hubbard Brook. Both inverse linear and inverse squared weightings were used, based on distances from Hubbard Brook to the estimated emissions centroids of the regions.

Multiple linear regressions were performed, using the stepwise mode and the meteorological variables listed above together with various emissions variables. The regression step selected for detailed consideration was either the one with the lowest standard error of estimate (highest adjusted R^2) or the subsequent step upon which the emissions variables entered the regression. Examination of residuals did not indicate autocorrelation to be significant, based on the Durbin-Watson statistic.

It was hypothesized that sulfate deposition at Hubbard Brook derived from two separate sources: aerosol which had been scavenged by precipitation, and sulfate formed in clouds. The aerosol portion was proportional to the concentration of ammonium ion (NH_4^+) in precipitation. Thus, the balance ($SO_{4eq}^{=} - NH_{4eq}^{+}$) would be a surrogate for the sulfate formed in clouds. An alternative estimate of in-cloud sulfate subtracted the calcium equivalent in addition ($SO_{4eq}^{=} - NH_{4eq}^{+} - Ca_{eq}^{++}$), recognizing that calcium is an important ion at Hubbard Brook, with a substantial downward time trend. However, since calcium is thought to be derived from suspended dust of coarser particle size that ammonium (sulfate), these two ions were not summed to represent aerosol derived from anthropogenic emission sources.

Table 1
Elasticities and 95% Confidence Limits

Relationship	Emissions Region	Summed Emissions	Linear Weighting	Squared Weighting
NO_3^- vs NO_x	State of NH	0.75±0.36	—	—
	Region I	0.25±0.45	—	—
	Regions I - II	0.70±0.85	0.89±0.62	0.70±0.51
	Regions I - III	1.41±0.58	1.17±0.69	0.84±0.56
	Eastern North America	1.12±0.61	1.03±0.57	0.88±0.51
	All U.S. + E. Canada	0.89±0.45	0.88±0.47	0.84±0.47
$SO_4^=$ vs SO_2	State of NH	0.05±0.30	—	—
	Region I	-0.27±0.44	—	—
	Regions I - II	-0.59±0.62	0.20±0.23	0.20±0.22
	Regions I - III	-0.42±0.84	0.30±0.32	0.24±0.26
	Eastern North America	-0.22±0.88	0.39±0.37	0.31±0.33
	All U.S. + E. Canada	0.52±1.00	0.59±0.71	±0.42±0.46
NH_4^+ vs SO_2	State of NH	0.60±0.52	—	—
	Region I	0.47±0.35	—	—
	Regions I - II	0.55±0.43	0.53±0.40	0.51±0.38
	Regions I - III	1.01±0.68	0.81±0.56	0.63±0.46
	Eastern North America	1.00±0.67	0.96±0.65	0.81±0.57
	All U.S. + E. Canada	2.77±1.70	1.94±1.19	1.18±0.77
$(SO_4^= - NH_4^+)$ vs SO_2	State of NH	-0.11±0.33	—	—
	Region I	-0.49±0.48	—	—
	Regions I - II	-0.91±0.68	0.11±0.26	0.10±0.24
	Regions I - III	-0.93±0.92	0.15±0.36	0.12±0.29
	Eastern North America	-0.73±0.96	0.21±0.42	0.17±0.37
	All U.S. + E. Canada	0.13±1.10	0.20±0.77	0.20±0.50

Table 1 presents preliminary results from this analysis, based on the following:

- Only data for which an ion balance was achieved within ±20% were retained.
- Annual rather than quarterly emissions were used (elasticities are slightly lower and less statistically significant when the quarterly values are used).
- 95% confidence limits are based on 15 degrees of freedom (t = 2.13).
- Emissions are aggregated by Federal region in three different ways: summation, weighting by the inverse distance from Hubbard Brook to the estimated centroid of emission, and weighting by the square of that distance.

Nitrate Ion

Statistically significant relationships were shown for almost all cases. In most cases, the elasticities are not significantly different from unity. There is no reliable indication as to which emission region provides the best fit, but there does seem to be a tendency towards an area larger than Federal Regions I and II.

Sulfate Ion

There are no statistically significant relationships between sulfate in rain at Hubbard Brook and SO_2 emissions for any of the regions considered. However, the weighted "all U.S. + Canada" cases nearly achieved significance with an elasticity around 0.5 (most likely value).

Ammonium Ion

Ammonium ion (taken here as a surrogate for sulfate derived from aerosol) shows statistically significant relationships with SO_2 emissions in all cases. Moreover, as the region increases in size, the elasticity increases, indicating a possible need to control over a wide region for maximum effectiveness. However, as with nitrate ion, it is not possible to indicate the optimum region for control with any degree of confidence.

Sulfate Less Ammonium

The difference between sulfate and ammonium equivalents is interpreted here as a surrogate for sulfate formed in clouds. No positive statistically significant relationships are seen. Since

it is difficult to conceive of a physically meaningful negative relationship, the weighted results appear to be the most credible and indicate no relationship between the two variables. This could indicate a situation of saturation, in which no more sulfate is formed if fresh SO_2 is supplied, and no less if SO_2 is reduced. Such a situation can only pertain locally, due to global mass balance considerations. Subtracting calcium in addition to ammonium ion did not materially change these results.

Figures 2 to 5 present plots of "seasonally adjusted" annual mean concentrations vs emissions. The "seasonal adjustment" procedure accounts for the effects of precipitation amount, monthly average temperature, and the seasonal periodic function (WAVE) by subtracting these effects as predicted by the regression equation, according to the following formula:

$$C_{seas.adj.} = C_{measured} - (K_1 \star PRECIP + K_2 \star WAVE + K_3 \star MAVTEMP)$$

where the K's are given by the averages of coefficients for several regression equations.

Figure 2 presents the time trends for sulfate in precipitation at Hubbard Brook and for the linearly (inverse) weighted SO_2 emission parameter, which includes all of the U.S. and Eastern Canada. After 1970, the trends are reasonably concordant, but before 1970 they are opposing. The $SO_4^=$ plot looks much like Fig. 4.14 in the Academy report, even though weighting by precipitation volume has not been done in Figs. 2-5.

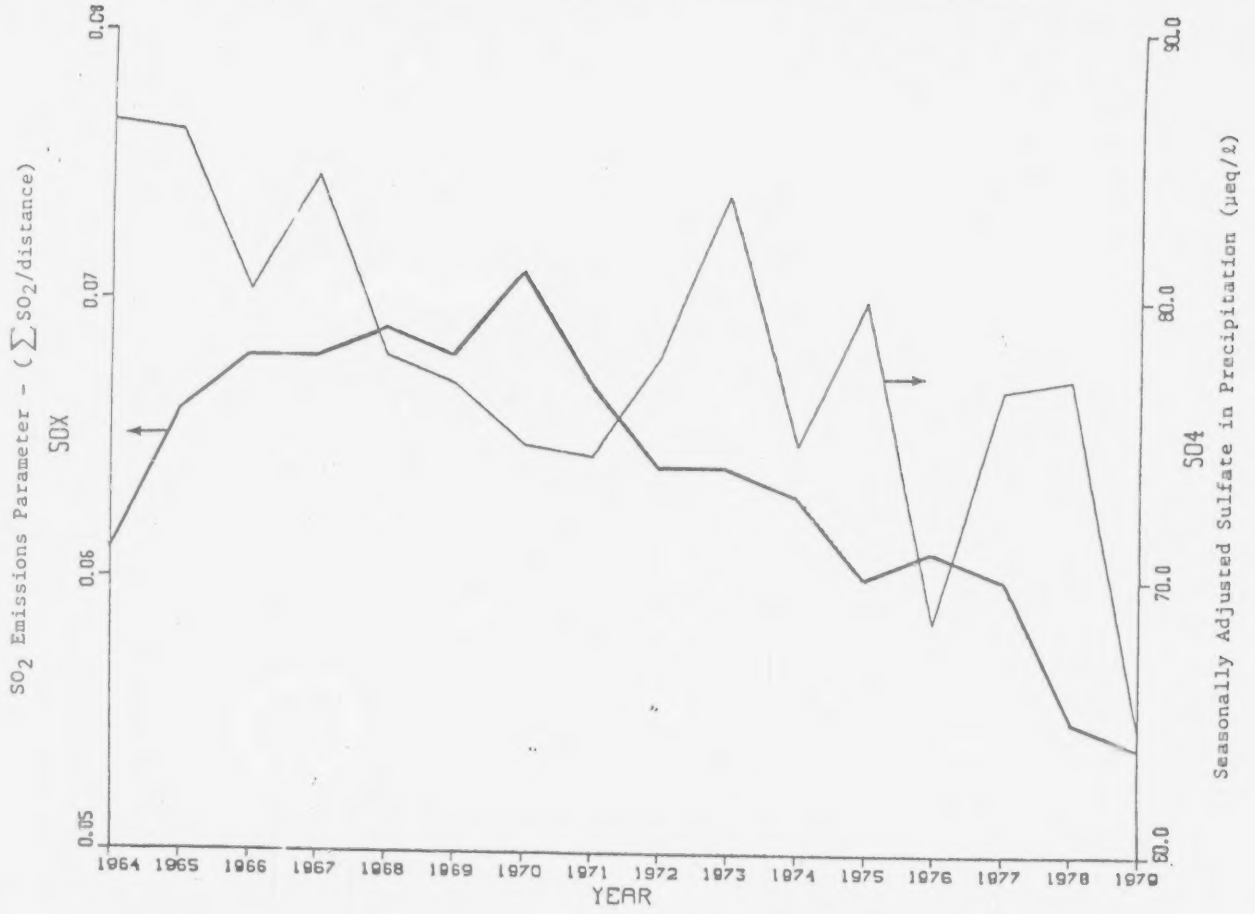
Figure 3 presents the cross plot, which indicates a weak (non-significant) positive relationship between the two parameters, with an elasticity of 0.35. An elasticity 1.0 line is also shown for comparison, and the two years used in the Academy report (1965 and 1978) are flagged. Note the very different impression created by considering just these two years.

Figure 4 presents a similar plot for ammonium ion, but in sharp contrast the data fit an elasticity $\simeq$ 1.0 line with a highly significant correlation ($P < 1\%$). Figure 5 presents nitrate data vs the linearly weighted (inverse) NO_x emissions parameter, which also results in a highly significant correlation, but a lower elasticity than obtained with the weekly sample analysis. The use of only 1965 and 1978 would provide a reasonable estimate in this case.

Conclusion

This analysis has indicated that at Hubbard Brook, proportional reduction might be expected in nitrate and ammonium deposition from the control of NO_x and SO_2 emissions, respectively. Considerably smaller benefits if any, would be expected for deposition of sulfate ion.

Figure 2. Time Trends for SO_2 Emissions and Precipitation Sulfate



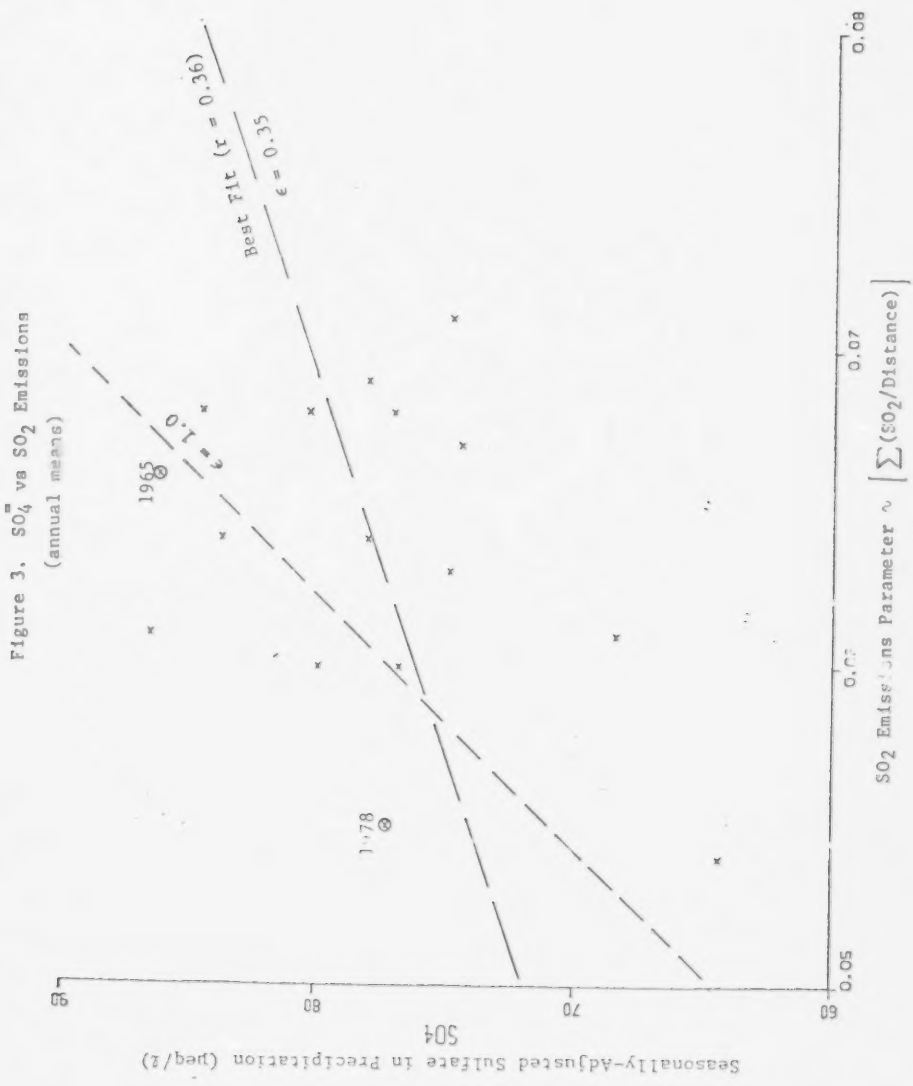


Figure 3. $SO_4^=$ vs SO_2 Emissions
(annual means)

Figure 4. NH_4^+ vs SO_2 Emissions
(annual means)

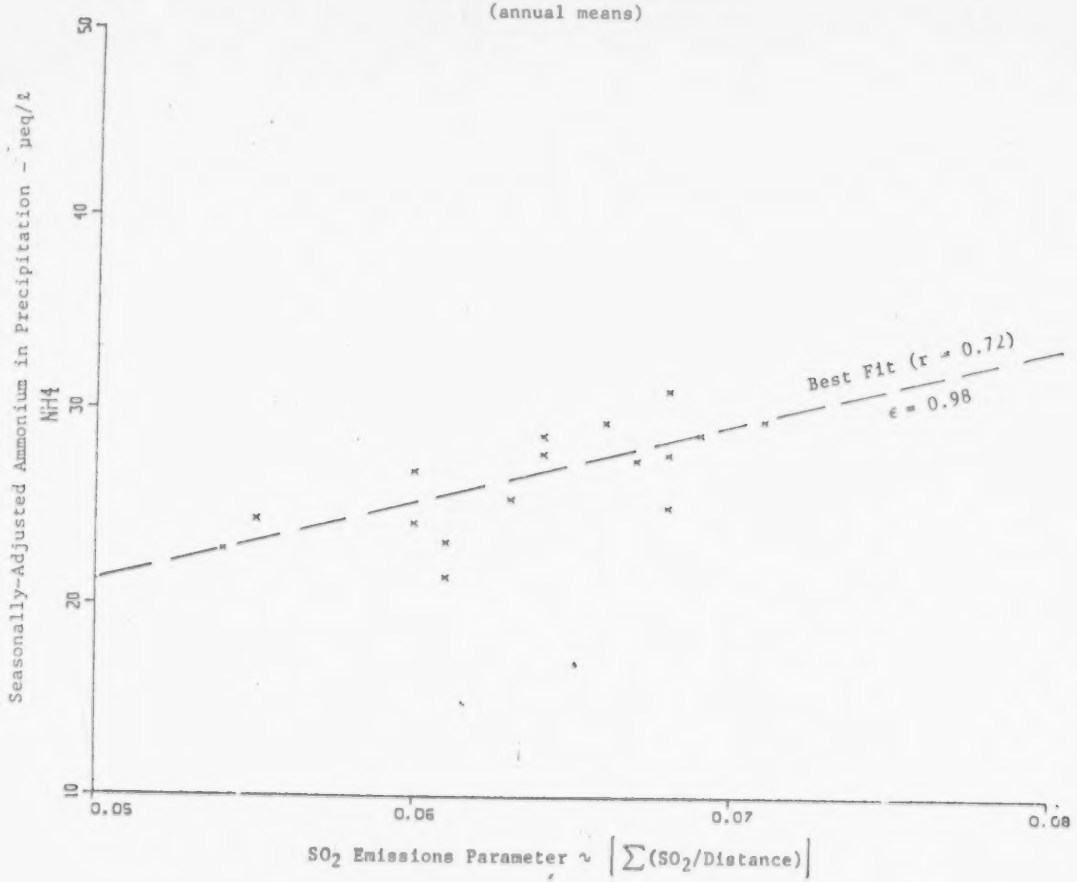


Figure 5. NO_3^- vs NO_x Emissions

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1. G.E. Likens et al., Biogeochemistry of a Forested Ecosystem, Springer-Verlag, 1977.

2. R.E. Munn et al., "A Climatological Analysis of the Hubbard Brook (NH) Precipitation Chemistry Data," Final Report to the U.S. Department of Energy, University of Toronto, Nov. 30, 1982.

3. Work Group 3B (Emissions, Costs, and Engineering Assessment), U.S.-Canada Memorandum of Intent on Transboundary Air Pollution, Final Report, June 1982.

4. U.S. Department of Energy, Energy Information Administration, Federal Energy Data System Statistical Summary Update, DOE/EIA-0192, July 1979.

5. Air Pollution Control Directorate, Environment Canada, EPS3-AP-80-5, January 1981.

6. Frank Vena, personal communication, Sept. 28, 1982.

Sulfate-to-Nitrate Ratios

The Academy report compares North American to European data for the molar ratio of (wet) deposited sulfate to nitrate in relation to the ratio of emissions, and argues that the greater uniformity of this deposition ratio in North America indicates the absence of nonlinearities in sulfate deposition processes, in contrast to the European situation.

What the report does not address in this regard, however, is the spatial distribution of the ratio of sulfur to nitrogen oxide emissions in relation to the ratio of deposition. The emissions ratio in Europe is not even mentioned (it is much higher than in the U.S.). This critique will attempt to develop these more detailed insights, for the U.S. situation, in order to test whether the Academy report's conclusions are still valid on a more detailed scale. The primary parameter to be examined is the "ratio of ratios," i.e., the molar ratio of sulfur-to-nitrogen wet deposition divided by the molar ratio of SO_2 to NO_x emissions.

Both spatial and temporal distributions are considered. For the spatial distribution, published deposition data are compared to the emissions data given in the Academy report. For the temporal distribution, the weekly samples at Hubbard Brook are compared to a year-to-year historical emissions data base which has been developed for that purpose (unpublished data).

In comparing Europe to North America, it must be remembered that almost all of the strong European sources are far from the regions where deposited sulfate begins to overwhelm deposited nitrate. In contrast, in North America, the source region is considerably larger and more varied in emission make-up. In the midwest, industrial SO_2 sources predominate, whereas in the east where fuel sulfur is strictly controlled, NO_x (transportation, home heating, etc.) reflects the higher population densities. Northern Canada should compare to Northern Scandinavia, however.

Spatial Distributions

Comparing Pack's[1] concentration isopleths to the emissions distributions in the Academy report (Fig. 4.18) we can look at the spatial distribution of $SO_4^=/NO_3^-$ in relation to emissions (SO_2/NO_x in molar units), rather than just in some undefined "region." For this purpose, transects were drawn along the estimated major and minor axes of the elliptical deposition isopleths, with peaks for both $SO_4^=$ and NO_3^- assumed to occur roughly at Steubenville, Ohio. Emissions ratios were plotted near the centroids of states, except for Illinois and Missouri which were averaged and taken at St. Louis in order to smooth the distribution.

Along the east-west transect, emission ratios show much more variation than the deposition ratios. The big emitting states have emissions ratios from 1.0 to 1.87, generally higher in the western portion of the transect. Deposition ratios are higher there also, decreasing to about 1.0 at the eastern boundary. If the deposition ratio is compared to the emissions ratio at the same location, the ratio of $SO_4^=/NO_3^-$ deposited to that emitted is of the

order of .0.6-0.7 in the source region, rising to 1.8 at the eastern boundary. Thus it is apparent that in the midwest, sulfate deposition is inhibited relative to nitrate and is transported to the east to be deposited there.

This point is even more obvious if a perpendicular transect is taken (generally) south from Steubenville to North Carolina. The deposition ratio remains at 1.1-1.3 almost all the way (there is an anomaly at the 30 μm/l $SO_4^=$ isopleth), whereas the emissions ratio in Virginia and North Carolina is at about 0.7. Since prevailing winds (if there is such a thing) are generally west-to-east, it may be prudent to translate the emissions transect to the west of the deposition transect to which it is to be compared. A distance of about 650 km was (arbitrarily) chosen for this purpose. On this basis, along the north-south transect, the ratio of $SO_4^=/NO_3^-$ deposited to that emitted 650 km to the west varies from about 0.6 to about 0.85, again with the lowest values in the source region reflecting a saturation condition. Going back to the east-west transect with a westward displacement of 650 km for emissions, we find a situation very similar to the south transect: ratios of $SO_4^=/NO_3^-$ deposited to emitted of 0.6 to 0.7 with the 0.7 value at the east coast. These data are plotted in Figure 1.

For a northern transect, it is necessary to use Canadian data, as for example shown by Semonin[2] as sulfate and nitrate deposition averages (mg/m^2) for combined CANSAP and NADP network data (monthly averages). The peak is shifted slightly to the north compared to Pack's data, but again, NO_3^- and $SO_4^=$ peaks roughly coincide. The molar ratio of deposition is about 1.0 in this peak region and begins to rise at about the location of the Sudbury smelters, reaching values of about 2.0-2.4 about 1100 km to the north. This trend is quite comparable to the European data shown in the Academy report.* Note that the very large SO_2 emissions from Sudbury are not felt immediately as increased wet deposition, especially not to the South** where NO_x emissions are highest. The emissions map (Fig. 4.18) indicates that in most of the Northern Canadian areas, sulfate should totally overwhelm nitrate (emissions ratios from 6.51 to 67.56), yet the deposition ratios do not differ greatly from the east-west transect in the U.S. (for coincident emissions and deposition). Deriving appropriate emissions ratios for this transect in Canada is thus quite difficult without recourse to meteorological models showing dominant storm tracks, in part because of the presence of numerous large water bodies to the south and west. Nevertheless, there is poor correspondence between deposition and emission ratios, indicating suppression of sulfate deposition.

A southwest-northeast transect into Atlantic Canada might be more appropriate than the previous due north transect. Similar results are seen by comparing emissions and deposition ratios in New Brunswick and Newfoundland (about 800 km apart), for example, as shown below.

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*It should be noted that if we are to rely on the spatial distribution of the European data, we should also rely on the temporal trends (which had been discounted in the Academy report).

**The perturbation in the N S transect curve in Fig. 1 at 500 km may reflect the presence of Sudbury, but the emissions ratio there is about 67, leading to a "ratio of ratios" of ~0 if non-coincident emissions are used.

	Deposition $SO_4^=/NO_3^-$	Emissions SO_2/NO_2	Dep/Emissions (Coincident)	Dep/emis. shifted NE
New Brunswick	1.61	2.62	0.61	
Newfoundland	1.95	1.51	1.38	0.74

Comparison on a coincident basis shows depression of sulfate deposition, which is reflected further downwind in Newfoundland as increased sulfate deposition. Comparing Newfoundland deposition to New Brunswick emissions (last column) shows sulfate deposition to be depressed, just as was shown in the U.S.

Returning to the Academy report, a presentation is made of the North-South gradient in deposited sulfate/nitrate ratio in Europe (1.1-2.0 below about 63° latitude rising to 5.7 in the far north), but no mention is made of the emissions ratio there. For the 11 countries (all in Western Europe) participating in the OECD Long Range Transport Study,[3] the ratio was 1.88. There is some reason to believe it could be even higher in eastern Europe, where high sulfur fuels are used and transportation sources of NO_x are less important. Thus, in Europe as well as North America, SO_4 deposition is inhibited relative to nitrate near source regions, resulting in excess sulfate deposition in more remote regions.

One final note: sulfate/nitrate deposition ratios tend to be elevated in urban areas, primarily because of higher sulfate in the winter than in rural areas, reflecting local source effects. (In general, nitrate deposition is not elevated in urban areas. Thus, in making spatial comparisons, the influence of urban areas must be excluded. It is also possible that this phenomenon reflects differing urban atmospheric chemistry as well, for example, the inhibition of ozone formation by local NO_x sources.

Temporal Trends

Thus far we have invoked only spatial trends to examine the sulfate/nitrate ratio question. Temporal trends can be invoked as well, since in general NO_x emissions have been rising, whereas after 1970, SO_2 emissions have generally declined. The Hubbard Brook precipitation chemistry data provide a sufficient record for this purpose.

First of all, there is a difference between the mean of the sulfate/nitrate concentration ratios and the ratio of the means (1.25 vs 1.06), reflecting a skewed distribution as well as correlation between sulfate and nitrate concentrations.* Since atmospheric chemistry acts on individual events and not on the means, the entire preceding discussion may be hopelessly clouded by the use of long term deposition averages.

Regression analysis was performed on the concentration ratios vs meteorological and various emission ratios (for successively larger regions), for years after 1965.* The only meteorological variables of significance were

*This figure is based on the years from 1966 onward; in 1964 and 1965, there were a number of low nitrate readings leading to sulfate/nitrate ratios as high as 120. However, Hubbard Brook personnel stand by the data as recorded.[4]

the monthly average temperature and the seasonal periodic function, both indicating an increase in the SO_4/NO_3 ratio in summer. The emissions ratio, was most significant when averaged over Federal Regions I and II, with an elasticity of 0.43 ± 0.35. When larger regions were used, elasticities increased somewhat, but statistical significance dropped. Correlations were quite modest; only about 15-16% of the variance in weekly samples was explained. The fact that elasticities were significantly less than unity is further indication of lack of strict proportionality between emissions and deposition. Thus, based on sulfate/nitrate ratios at Hubbard Brook, there is no assurance that a change in emissions ratio would be reflected in a proportional change in deposition ratio.

Conclusions

The Academy report concludes that sulfur and nitrogen atmospheric and deposition processes are quantitatively similar since both emissions and deposition ratios are about 1.0 when averaged over a large region. They invoke a comparison to the European deposition ratio distribution to show that Europe and North America display different atmospheric chemistry situations.

This review has shown that in North America, sulfur deposition is depressed relative to nitrogen in source regions, rising to show an excess in more remote (or lower emissions density) regions. The Academy's use of regional averages obscures this fact. This depression of sulfur deposition may indicate a saturation phenomena in regions of large sulfur emissions, which would constitute a significant non-linearity in the system. In addition, when the European sulfur-to-nitrogen emissions ratio is considered, no important differences appear between the two continents.

Thus, in contrast to the Academy's findings, the data on sulfur/nitrogen ratios support the presence of significant non-linearities in the system, which would result in a disproportional relationship between changes in emissions and changes in deposition.

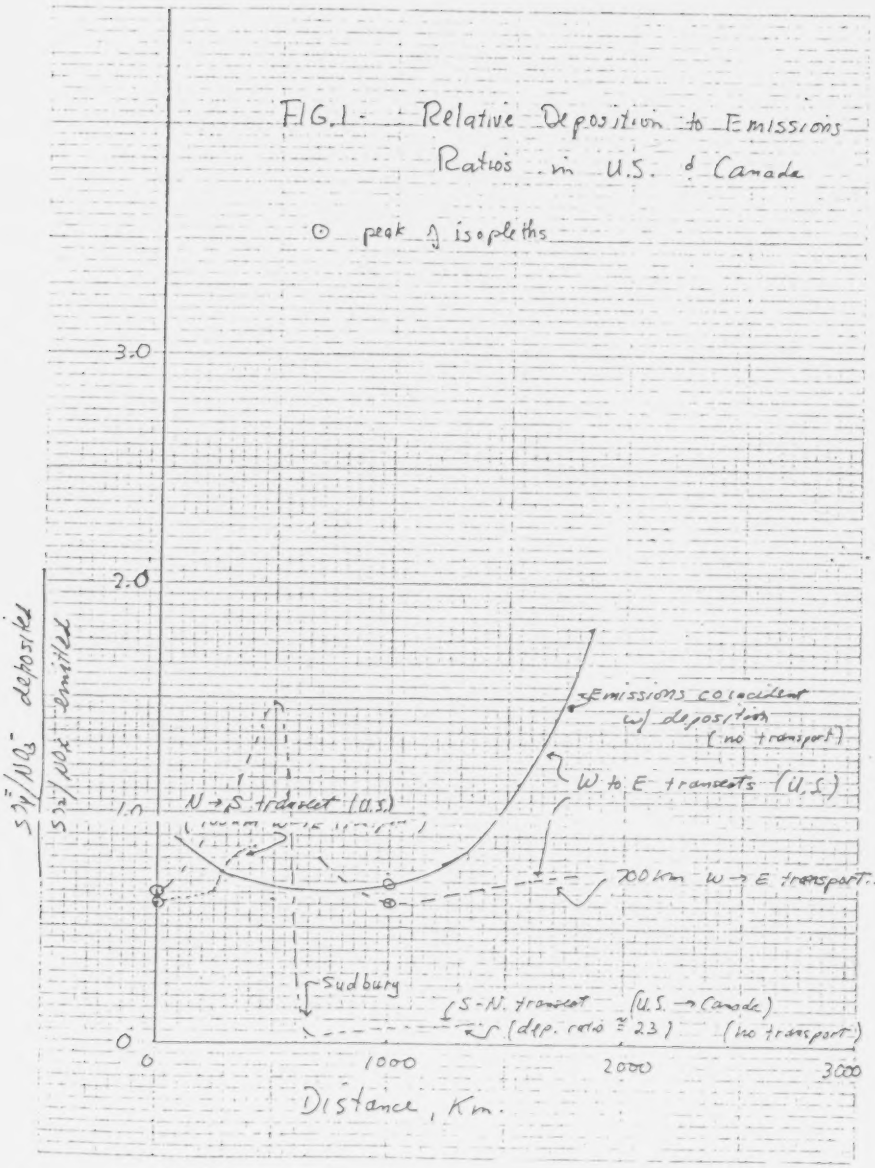
FIG. 1 - Relative Deposition to Emissions Ratios in U.S. & Canada
⊙ peak ∿ isopleths
3.0
2.0
1.0
0
0
1000
2000
3000
Distance, Km.
Emissions coincident w/ deposition (no transport)
W to E transects (U.S.)
N → S transect (U.S.)
700 km W → E transport
Sudbury
S-N. transect (dep. ratio ≅ 23)
(U.S. → Canada) (no transport)

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2. Semonin, R.G. "Seasonal Precipitation Concentrations and Depositions for North America from the CANSAP/NADP Networks," AIAA 82-0334, January 1982.

3. Organization for Economic Co-Operation and Development. The OECD Programme on Long Range Transport of Air Pollutants, Measurements and Findings, Paris, 1977.

4. Likens, G.E., personal communication, Aug. 1, 1983.

The Committee on Atmospheric Transport and Chemical Transformation in Acid Precipitation of the National Academy of Sciences in their recent report state that "there is no evidence for a strong nonlinearity between long-term average emissions and deposition," (Summary page 7). They admit that the conclusion is clouded by uncertainty (p. 9) due to "the limited amount and uncertain quality of the data, the natural variability of atmospheric processes, and the lack of firm understanding of the physical and chemical processes involved." Beyond these disclaimers, there are a number of questions that can be raised concerning the basis for the NAS conclusions. These points need resolution before the conclusions are used for the setting of emission control policies.

1. ONLY WIDE-AREA, LONG-TERM CONCLUSIONS ARE DRAWN

The NAS committee looked at long-term data only; they did not address the short-term (event) or the spatial scale of less than a few hundred kilometers. The million-square kilometer so-called mixing that they discuss occurs only as an artifact of averaging over long time periods.

On the other hand, it is, no doubt, true that if a large enough area is considered, the acid deposition response to emissions will be linear. The real question is "On what scale is the system linear?"

2. THE USE OF HUBBARD BROOK DATA IS QUESTIONABLE

The deposition concentrations at Hubbard Brook are somewhat lower than those in the region of highest concentrations (by almost a factor of two).

The very nature of mechanisms that can cause nonlinearities (i.e., limited amounts of oxidant, competition among species, pH-induced limitations on mass transfer, etc.) means that, at first approximation, the effect will be most evident where the concentrations are the highest. However, because of the fact that the system will appear linear if averaged over a large enough region, an opposite effect

is liable to be seen where lower concentrations occur far downwind of the high source and concentration regions. For example, a lack of sufficient oxidant in a region would allow unoxidized SO_2 to be vented from a storm and made available farther downwind. Hence, an increase (or decrease) in emissions might result in a smaller than proportional increase (or decrease) in concentrations near the regions of high emissions; thus, a proportionally greater (or lesser) amount of unreacted primary pollutant will be available to be deposited downstream. Following this line of reasoning, it is quite possible that Hubbard Brook could be in a location where ambiquous results on linearity would be obtained. The attached figure illustrates the point.

3. INSUFFICIENT ATTENTION IS GIVEN TO IN-CLOUD CHEMICAL REACTIONS AS A POSSIBLE SOURCE OF NONLINEARITY.

There is a large amount of admittedly inferential evidence that a major portion of precipitation acidity derives from in-cloud oxidation. Quite a number of authors have put forth this point of view (Newman, 1979; Scott, 1982; Lazrus, et al, 1983; Oppenheimer, 1983; etc.). Hence, the effort to discredit the model of Rodhe, et al, (1981) might well be moot; the Rodhe treatment of in-cloud oxidation is quite simplified and does not seem to have been improved in the calculations reported in the NAS report. The report discusses how H_2O_2 production might be suppressed in a NO-rich polluted atmosphere and how it could be rained out early in a storm leaving unreacted SO_2. They recognize that this could be the source of nonlinearities, but note the lack of an adequate data base to test the possibility. The report states that the data "suggest that concentrations of (oxidant in cloud water) may be sufficient in eastern North America for complete oxidation of SO_2 except, perhaps, in winter." There are a number of photochemical calculations that would indicate that the H_2O_2 concentrations are several times too low. At best, the point is arguable; it might just be that the NAS report is incorrect.

4. "LACK OF EVIDENCE FOR STRONG NONLINEARITIES" IS NOT A PROOF OF LINEARITY.

Stating that "no evidence of strong linearity" could be found is far from being a logically sound proof of linearity.

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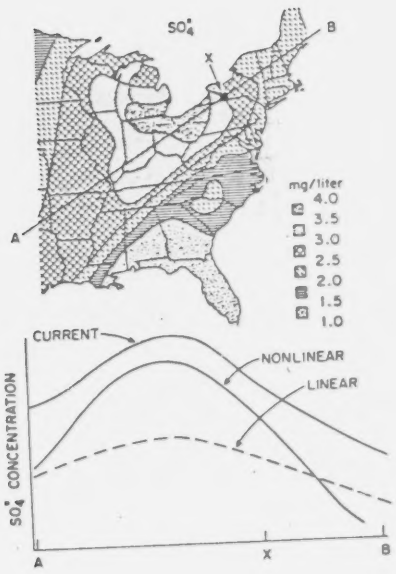

Fig. 1. Top--Observed 1979 average concentration of $SO_4^=$ in precipitation (after Wilson and Mohnen, 1982). Bottom--Current concentration along cross section A-B and hypothetical concentrations that would result from an emission reduction if the response were linear or nonlinear.

Review BNL-C

Comments on
"Acid Deposition - Atmospheric Processes in Eastern North America -
A Review of Current Scientific Understanding,"
National Research Council.

Three major issues are considered and appear to serve as foci for the material reviewed in the report. As stated on pages 6-9, these issues are (1) "applicability of models to decisions on control strategies," (2) "non-linearity" and, (3) "influences of local and distant sources."

I. Models:

The authors have applied a healthy degree of skepticism and insight in arriving at their conclusions regarding the usefulness of models. To paraphrase the report (pp 148-149) ..... a great deal of research is required before the acid deposition problem can be described with precision by models. It is fairly clear that in this statement the usefulness of models is judged relative to the needs of policy makers, not to the needs of research scientists. The use of models, even complicated ones, by the latter group, should be judged by different standards. Although models cannot be used to make a priori predictions of many of the important features of the acid deposition problem, they can be used to good advantage by the research community in designing experiments, interpreting data, performing controlled computer experiments on the environment and, in general, increasing our understanding of how the various pieces of a complex problem interact with each other.

II. Nonlinearity:

An unsupported conclusion is reached regarding the relation between emissions and deposition. "It is ..... improbable that the oxidation of SO_2 is sufficiently hindered by a lack of oxidant to cause a disproportionately small

reduction in sulfate concentrations as a result of a given reduction in SO_2 emissions." (p 9). The "evidence" in the text for this statement derives from a comparison of deposition at Hubbard Brook with emission, from consideration of the gas phase oxidation chemistry of SO_2 and from the sulfate to nitrate molar ratio in precipitation as compared with emissions. This evidence, some of which can be interpreted in different ways, as described below, does not support the stated conclusion that deposition is linearly related to emissions. At best, one could say, "Direct evidence of a strongly nonlinear relationship between wet deposition of sulfate and SO_2 emissions is limited to extensive historical data in Europe." (p 139). Specific comments on the "evidence" against nonlinearity follows:

- p 42, also p 84. "In theory, the amount of H_2O_2 formed in gas-phase . . reactions and taken up in cloud water and precipitation, is sufficient to oxidize a large fraction of S(IV)." Many photochemical calculations of the PBL predict a H_2O_2 concentration of a few tenths to approximately one ppb (Schiavone and Graedel, 1981; Rodhe, et al, 1981; Hov, 1983). In order to account for the composition of summertime precipitation in the northeastern U.S., several times this amount of oxidant and SO_2 is required. It is certainly possible that future calculations or experiments will show that $[H_2O_2] > [SO_2]$, perhaps as a result of an in-cloud or free tropospheric source of H_2O_2. It is misleading, however, to imply that a large enough oxidant supply has been identified.

- p 50. "The most important attachment process under most in-cloud conditions is undoubtedly nucleation." Judging from a following sentence, diffusional attachment (of gases and particles) appears to be a subset of "attachment."

Either there is a semantic difficulty or the role of gas phase SO_2 and HNO_3 in acidifying precipitation is seriously understated.

• p 80. The criticism of the nonlinearity produced in Rodhe's model is supported by many other experimental and theoretical studies (see, e.g., Miller, 1978; Hov and Isaksen, 1981). Although it appears likely that the gas phase oxidation of SO_2 is linear, there is no assurance that the aqueous phase oxidation is likewise linear. It is a deficiency of the main text (Chapters 1-5) that the relative amounts of gas and aqueous phase oxidation are not treated. Estimates of the relative importance of these processes have been made based upon the isotopic composition of precipitation (Holt, et al, 1981), the abundance of aerosol SO_4^{2-} (Newman, 1979; McNaughton and Scott, 1980), and a comparison of the acidity of precipitation and aerosol (Newman, 1979; Scott, 1982). At this point, the evidence, though largely inferential, points strongly to the in-cloud oxidation process as being the largest contributor of SO_4^{2-} to precipitation (Oppenheimer, 1983). The concern is that the aqueous pathway is the most significant and that it is nonlinear due to either pH inhibition or oxidant limitation.

• p 120-123. Comparison of Hubbard Brook data with emissions. It is misleading to base this comparison upon two years of monitoring and emissions data, namely 1965 and 1978. Emissions have a broad maximum around 1970 which is not reflected in precipitation composition. The two sets of curves, in fact, look sufficiently dissimilar such that one could argue that there is a complicated nonlinear relation between emissions and deposition.

• p 135. The NO_3^- to SO_4^{2-} ratios do not justify the conclusion that there is sufficient oxidant for all or most of the SO_2 within eastern North America. It admittedly is more straightforward to assume that equal deposition and emission ratios imply a high removal fraction for both substances than to arrive at an

explanation for why the average removal fractions are both equal but substantially less than one. The proposed high removal efficiency of NO_x is based upon the rapid formation of HNO_3 and the high solubility of HNO_3. A high NO_x wet removal efficiency is, however, not in accord with summertime, air quality observations in moderately polluted areas which indicate

$$\frac{SO_4^{2-}}{SO_2} > \frac{HNO_3 + NO_3^-}{NO_x} .$$

Perhaps HNO_3 is rapidly removed by dry deposition thereby making it unavailable for wet deposition. A NO_3^- to SO_4^{2-} molar ratio in precipitation equal to emissions could then occur without requiring efficient scavenging of S compounds.

• p 137. The data in Table 4.9 reflects the different average heights of NO_x and SO_2 emission sources. In general, during the nighttime $NO_x > SO_2$. This occurs, in part, because low level NO_x emissions are trapped above the boundary layer.

III. Source Attribution:

The text contains a good description of the monitoring evidence to the effect that receptor areas are impacted from many source regions, sometimes simultaneously. The complexity of the source attribution problem is conveyed as are the hopes and shortcomings of several diagnostic methods. The notion of strong mixing is, however, considerably overstated due to the authors' reliance on long term (monthly) average monitoring data.

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Oppenheimer, M. (1983). The relationship of sulfur emissions to sulfate in precipitation. Atmos. Environ. 17, 451-460.

Rodhe, H.F., Crutzen, P. and Vanderpol, A. (1981). Formation of sulfuric and nitric acid in the atmosphere during long range transport. Tellus 33, 132-141.

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Review BNL-D

Review of National Academy Report on Acid Deposition

This report focuses upon the relationships between emissions of acid precursors and the deposition of acidic substances. The stated purpose is "to assess the current state of scientific information that can be marshaled to describe those relationships in the hope that our assessment will be valuable to decision makers in the government and in the private sector" (pp. 1-2).

Overall, the report exhibits a number of positive qualities, but in my judgment it is seriously flawed. On the positive side, it delineates well the scientific issues that confront the regulatory sector. The red herring of natural sources is readily dispensed with (p. 3). The scientific issue is thus drawn as establishing the relationships between anthropogenic emissions and deposition. The report also defines the sub-issues of nonlinearity and distant vs. local sources. The report draws the conclusion that "evidence exists for long-range transport of pollutants leading to acid deposition", but cautions that "the relative contributions of specific source regions to specific receptor sites currently remain unknown" (p. 5). The report points out gaps in knowledge requisite to establishing such contributions and suggests pertinent laboratory, field, and modeling studies to better define such relationships. The report concludes that at the present level of understanding "we cannot objectively predict the consequences for deposition in ecologically sensitive areas of changing the spatial pattern of emissions in eastern North America, such as by reducing emissions in one area by a larger percentage than in other areas" (p. 11). In my judgment, the report makes an adequate case for the above conclusions. However, as I argue below, the report might well have offered some guidance, particularly with respect to the latter question.

On the negative side, I submit that the report is deficient insofar as it focuses, almost entirely, on long-term average (e.g., annual) deposition, to the exclusion of consideration of information that has been gained from shorter-term measurements. While such a focus may be appropriate from the perspective of ecological concerns and/or from a regulatory perspective, a wealth of detail is lost that is pertinent to the mechanism of acid deposition, and pertinent, therefore, to the stated subject of the report. The focus on long-term relationships between emissions and deposition leads to a number of conclusions that either are erroneous or subject to misinterpretation or, alternatively, are so narrowly drawn as to border upon the self-evident. I call particular attention to the following:

(a) "Pollutants are being thoroughly mixed over a region with linear dimensions up to 1000 km" (p. 9). This conclusion derives from the claimed near-constancy (in space and time) of the ratio of long-term average sulfate and nitrate concentrations in precipitation and the near equality of this ratio to the ratio of the oxides in emissions. The statement is patently incorrect if we take "thoroughly mixed" in the usual sense of "exhibiting uniform concentration over the volume in question, at a given instant or

over a given period of time." Evidently, the authors mean simply to say that the ratio of long-term average atmospheric concentrations is uniform spatially. In any event, the statement as it stands is erroneous and certainly detrimental to conveying the authors' point.

(b) "There is no evidence for strong nonlinearity in the relationships between long-term average emissions and deposition" (p. 7). Presumably the authors mean to say here, "There is no evidence for strong nonlinearity in the relationships between long-term average emissions and long-term average deposition." Accepting that, the statement summarizes the search for such a relationship in available, long-term records of precipitation concentration. As it turns out, there is essentially only one station that meets the authors' criteria for such records of acceptable quality, and the authors claim to discern no such relationship for that station. While that may be argued (and I am sure it will be), restricting the discussion to long-term averages excludes a wealth of evidence from short-term measurements (event or sub-event) that is demonstrative of mechanistic nonlinearities, the understanding of which is crucial to the description of the source-receptor relationship in acid deposition.

(c) "If we assume that all other factors, including meteorology, remain unchanged, the annual average concentration of sulfate in precipitation at a given site should be reduced in proportion to a reduction in SO_2 and sulfate transported to that site from a source or region of sources" (p. 10). This statement is so highly restrictive as to be essentially useless. Strictly, it deals only with the relationship between annual average concentration of sulfate in precipitation at a given "site" and, presumably, annual average atmospheric SO_2 and sulfate at that "site," under the assumption that "all other factors, including meteorology, remain unchanged." Even granting the validity of the statement as it stands, the statement is all the more important for what it does not treat, i.e., (1) the relationship between emissions and annual average atmospheric concentrations at sites remote from emissions, and (2) the factors which are assumed to remain unchanged. While it might be hard to quarrel, on the basis of annual-average measurements, with the statement as it stands (although I can think of at least three mechanistic reasons why it might not be expected to hold in the short term and can cite, as well, any number of studies to that effect), I am concerned that the statement may be misconstrued as relating deposition to emissions, particularly insofar as it appears under the heading "Implications for Emission-Control Strategies," and immediately follows the statement that "analysis of observational data provides guidance for assessing the consequences of changing SO_2 emissions for wet deposition of sulfate."

(d) "Assuming that all other emissions and conditions remain unchanged, we would expect that if the molar ratio [of SO_2 to NO_x] in emissions in eastern North America were changed by changing SO_2 emissions, a similar change would occur in the ratio of sulfate to nitrate in wet deposition" (p. 10). The authors then go on to qualify the statement, indicating that "because the analysis is based on spatial distributions, its applicability is limited to circumstances in which the spatial distribution of emissions is not changed." Here, as above, I am concerned about possible

miscontruction of the intent of the authors' statement. I am sure that some will see in this statement a policy implication that if sulfur oxide emissions are to be reduced, they should be reduced in such a way as to preserve the existing spatial distribution. On the other hand, I would foresee others who would take statement (a) above, that "pollutants are being thoroughly mixed over a region with linear dimension up to 1000 km," as suggesting that reduction in SO_2 emissions anywhere within the region will be equally effective in reducing annual average deposition of sulfate at all other points in the region. I submit that the proper strategy for emissions reduction to protect ecologically sensitive regions lies somewhere between these two extremes. Unfortunately, the report provides very little guidance to the policy makers who must formulate such a strategy.

In addition to the foregoing general comments, I have a number of comments relative to the linearity-nonlinearity issue. But first we must examine why linearity-nonlinearity is of interest at all. Clearly, if the geographical region is taken sufficiently large, then the long-term total deposition of SO_2 and $SO_4^=$ integrated over this area is linear in emission of SO_2 in this area, and for that matter equal. The question of nonlinearity in long-term deposition is therefore inherently a question of whether the scale of transport from source to receptor is influenced by the emission source strength. Clearly also, any negative departure from linearity in a given receptor region must be compensated by a positive departure from linearity in some other receptor region or regions. Thus, it is evident that the entire question of linearity-nonlinearity is deeply imbedded in the larger question of the scale of transport of the pollutants of concern. This point is not well brought out in the report. The failure to appreciate this point may be responsible for some of the confusion in the report over the question of nonlinearity.

Some specific comments on the treatment on nonlinearity follow.

(1) For all the care given to the definition of linearity (pp. 72-75), the report still seems to confuse linearity with proportionality. Thus, (p. 116): "The central question is whether the data [obtained from monitoring networks] can be used to judge whether deposition is linearly or nonlinearly related to emissions.... If the relationship is linear, a change in emissions would be reflected in a proportionate change in deposition; if it is nonlinear, the change in deposition, if any, would be disproportionate." The second statement above is at best loosely phrased and at worst flat out wrong. It could be corrected as follows: "If the relationship is linear, a change in emissions <u>from a given source (or set of sources)</u> would be reflected in a proportionate change in deposition <u>due to that source (or set of sources)</u>," and similarly for the second clause. The addition of the logical connection between the source and the receptor is, in my judgment, central to the entire question of acid deposition, and ultimately to the question of control strategy. However, setting that aside for the moment, it is disappointing to say the least to see propagation of such confusion in this forum.

Going beyond definitions, we see that the foregoing misconception is carried over into the discussion of the model runs by Samson (p. 76). Here, calculations are compared with and without the contribution of a "background" source. The report states that the shift from the solid to the dashed sets of curves depicted in Figures 3.1 and 3.2 is "the result of eliminating the trivial source of nonlinearity arising from the background source, term B in the equation, $c = kS + B$." This result is indeed trivial but it is not a nonlinearity and in any event is an observation that should hardly warrant discussion. The only reason that such a discussion can possibly have persisted into the final report is an inherent lack of understanding of the linearity-nonlinearity concept.

One further amusing (or saddening) point is the passing reference in the same paragraph to Figure 3.2 depicting "a result that is essentially linear (with proportionality constants less than unity)." The point of figures such as 3.1 and 3.2 is that any departure from the one-to-one line of percent $\Delta SO_4^=$ concentration vs. percent ΔSO_2 (concentration or emission), sulfate concentrations from other, unchanged sources having first been subtracted out, is an inherent nonlinearity. In fact, the 24- and 48-hour graphs represent a departure from linearity of ca. 30%, potentially very important in the context of control strategies. This is not to argue that the model calculations are correct, but only that they are incorrectly interpreted.

(2) The report argues at some length (pp. 75-81) that model calculations of Rodhe et al. suggesting nonlinearity may be an artifact as a consequence of the incorporation in the model of a radical termination step involving SO_2, whereas the actual reaction may well be a chain propagation step: "The participation of reaction (3.1) results in a direct nonlinear feedback into the SO_2 oxidation mechanism, while reaction (3.2) does not seriously perturb the concentration of the hydroxy radical" (p. 78). This argument seems flawed by the following reasoning. We note (p. 80) that in the simulation studies of Calvert and Stockwell, only about 10% of $HO-HO_2$ radical termination occurs through the $HO-NO_2$ reaction, for the quite high NO_x concentration of 100 ppb (and less for lower NO_x; indeed, the report suggests a curious quadratic dependence). In any event, we note that the rate constant of the $OH-NO_2$ reaction is about tenfold that of the $OH-SO_2$ reaction, so that for 100 ppb SO_2, only 1% of radical termination would be due to SO_2 (again less for lower SO_2). Thus, the possibility of any significant influence on radical concentration due to SO_2 termination reactions would appear to be ruled out.

(3) The discussion of nonlinearity in SO_2 deposition due to aqueous-phase processes (cloud, rain) is short (1.5 pages; pp. 81-82) and superficial. Reference is made essentially only to the H_2O_2 oxidation mechanism of SO_2 oxidation, focusing principally on the limiting reagent process. No discussion is given at all of the pH dependence of SO_2 solubility and aqueous reaction kinetics, which would be inherently nonlinear insofar as H_2SO_4 (and to a lesser extent SO_2 itself) would influence cloudwater pH. This process has been the subject of much research over the past decade (as summarized in Appendix A, pp. 178-193), but little of this research finds it way into the discussion in the text. Other types of processes leading to nonlinearity (e.g., non-constant nucleation efficiency as a function of aerosol loading) are also not treated.

In summary, the relationship of the linearity-nonlinearity issue to the larger problem of scale of transport and control strategies does not appear to be adequately recognized. Moreover, the details of the discussion of nonlinearity are superficial and flawed, and little recognition is given of the importance of the fact that nonlinearities, as exhibited in short-term phenomena, can influence the source receptor relationship. For these reasons, little import should be given to the conclusion of a lack of evidence for nonlinearity in the relationship between long-term average emissions and deposition.

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August 29, 1983

Dr. Bernard Manowitz
Chairman, Department of
Applied Sciences
Brookhaven National Laboratory
Upton, New York 11973

Dear Dr. Manowitz:

I am writing to you in your capacity as Chairman of the National Laboratory Consortium of the Interagency Task Force on Acid Precipitation. In response to your request we have reviewed the National Academy of Sciences' report entitled "Acid Deposition: Atmospheric Processes in Eastern North America".

The attached comments were provided by a review team comprised of research scientists from several divisions within the Laboratory and from a local institution we collaborate with in several research activities. This team of scientists collectively presents considerable experience in the field of atmospheric deposition. The reviewers consisted of (A) S. E. Lindberg, a chemist with 7 years' experience and over 40 publications in the area of air pollution research with special emphasis on forest canopy/atmosphere interactions and G. M. Lovett, a biologist with 5 years' experience and 7 publications in the area of atmospheric deposition to plant canopies and chemical interactions between plants and the atmosphere; (B) F. C. Kornegay, a meteorologist with 8 years of experience and over 20 publications in the area of air quality impacts of energy production and utilization, with special emphasis on atmospheric turbulence and diffusion, and (C) B. B. Hicks, a meteorologist with over 20 years of experience and 70 publications in the areas of precipitation scavenging, dry deposition, and atmosphere/surface exchange phenomena. We believe these people are highly qualified to review the National Academy of Sciences' report.

Our reviewers express concern about inadequate discussion of uncertainty levels associated with the assumptions upon which conclusions of the NAS report are based. We believe this is an important point

because of the natural tendency of nontechnical readers to develop a position or make a decision without regard to the uncertainty associated with the conclusions of a given study. You will also note an objection to the chemical, rather than a meteorological, focus of the nonlinearity issue.

Please let me know if you need additional information.

Sincerely,

Chester R. Richmond

Chester R. Richmond
Associate Director for Biomedical
and Environmental Sciences

CRR:ja

Attachments

cc: S. I. Auerbach
B. B. Hicks
F. C. Kornegay
S. E. Lindberg
G. M. Lovett

The NAS Committee on Atmospheric Transport and Chemical Transformation in Acid Precipitation was given the difficult task of addressing an exceedingly complex issue in an objective scientific manner. We feel that the committee has presented a reasonable "best guess" regarding the linearity between emissions and deposition. The challenge now is for the scientific community to refute, substantiate, or provide reason to alter this conclusion. For this reason, the conclusions in this document should not be used for establishing ultimate air quality regulations, but can be very useful in the initial development of policy.

The following points are made in support of this contention and discussed further below: (1) uncertainty is not clearly expressed; (2) conflicting European data is summarily dismissed; and (3) many potential problems with the Hubbard Brook data are not considered in detail.

1. Uncertainty - The degree of uncertainty in many of the calculations and assumptions used to conclude that there is not a strong case for nonlinearity is buried throughout the chapters and appendices. In some cases, uncertainty has been quantified, while in others it has not. When the reader considers all of the sources of uncertainty involved, he is left with the unmistakable conclusion that the 1:1 relationship between emissions and deposition attributed to the report by the press is highly uncertain.

2. European data - Not enough time has been spent in analysis of the European network data which provides evidence for nonlinearity. The European data is far more extensive in both space and time than the single Hubbard Brook data set on which so much emphasis has been placed. The reasons for dismissing these data may also apply to the Hubbard Brook data as well (see below). The Hubbard Brook data set is only one of four wet deposition data sets cited in the report. The others were dismissed because of "too much scatter," "uncertain quality control," and because they are "difficult to interpret." Perhaps all of these data warrant further analysis.

3. Hubbard Brook data set - The analysis of the Hubbard Brook data, on which the important conclusion on linearity is primarily based, leaves some questions in the minds of the reader. The contradictory European data on sulfur deposition are dismissed as being "somewhat uncertain" because of changes in analytical and sampling techniques during the period of observation. No attempt is made to convince the reader that the Hubbard Brook data are not subject to the same uncertainties. Did the site operators and analytical techniques for Hubbard Brook change between 1964 and 1978? For instance, ion chromotography has supplanted automated colorimetry as the technique of choice for analysis of sulfate and nitrate in water samples--did such a change occur at the laboratory analyzing Hubbard Brook samples?

   Another point involves the use of annual average trends in NO_3^-, SO_4^{2-}, and the SO_4^{2-}/NO_3^- ratio in precipitation. Galloway and Likens (Atmospheric Environment 15:1081-1085, 1981) reported that the trends in deposition at Hubbard Brook were seasonal, with NO_3^- increasing in the summer and SO_4^{2-} decreasing in the winter. Thus, the SO_4^{2-}/NO_3^- ratio decreased year-round, but for different

reasons in different seasons. It seems necessary to show that the seasonal patterns of deposition match the seasonal patterns of emissions, rather than analyzing the composited annual data as done in Chapter 4.

The fact that the Hubbard Brook data is bulk precipitation clouds the entire analysis. Keeping in mind that this is the only type of long-term data available, one needs to consider its limitations. Bulk samples contain an unknown, but generally small contribution of dry deposition, particularly at sites distant from local sources and particularly for gases. Yet, the Hubbard Brook researchers have estimated that dry deposition of SO_2 can be a significant source of deposited S, which is not collected in bulk samples. Recent data on the possible importance of dry deposited HNO_3 vapor add further evidence to support the statement in the report that dry deposition may be comparable to wet. The fact that the case for linearity has been based on samples containing primarily wet deposited material further emphasizes the uncertainty of the conclusions.

28-916 0 - 84 - 63

Review ORNL-B

SUBJECT: Acid Deposition Document Review

I felt that the document was a farily complete review of pre-1982 acid rain research. The material covered was extensive, but I certainly noticed bias in the writing towards conventional modeling studies. Little mention of other studies, such as ANL's work on examining the potential formation mechanism of sulfates in precipitation, was made. My major criticism of the document, however, is that modeling is in fact denounced as a regulatory tool in the summary (p. 5), but when one muddles through the text, the bottom line on p. 83 reads ".. we advise caution in projecting changes in deposition patterns that result from changes in emissions of precursor gases." I feel that such a summarizing statement in the modeling chapter is a de facto endorsement of the continued reliance on LRT models, an endorsement that has no basis in scientific fact.

Specific Comments

Page	Comment
7	The conclusion of no evidence of non-linerarity is not borne out by the data. More recent work, particularly by McLaughlin, indicates the importance of local sources.
11	I strongly support the need for less modeling and more direct field work. However, no acknowledgement of past long distance field work was given, and no recommendations as to best techniques were made.
59	If the gross statistical codes are as good as the transport codes, this is overwhelming evidence that the transport codes are missing the basic physics.
67	The reference to Fig. 2.5 indicates uncertainty to the author. To me, it indicates a total inability to determine where material goes, which implies an inability to provide any useful insight into the problem.
83	I find the conclusion of advising caution to be far too weak. As I read the chapter, and the data that are referenced, my conclusion would be that present LRT models are of no value in a regulatory process, and should not be used in any way, shape, or form.
111	I've never seen so many "ifs" and "thens" in a paragraph in my short life as in the first paragraph. Also, the reliance on such a tenuous assumption, when other studies have been unable to correlate SO_2 and SO_4 concentrations, is not in keeping with a national summary.
119	If no changes in long-term precipitation pH in the only decent record can be detected, and if no change in H^+ deposition can be noted, where is the problem?

For my summary, I would repeat that I, perhaps being overly sensitive, detect an unpalatable bias in the document. I also feel that the report is mostly complete, but serves only as a chronicle of work done to 1982, without shedding much light on what to do next.

Review ORNL-C

I regret that I have not had time to review the NAS Acid Deposition Document as thoroughly as I would have liked, nor am I going to have time during the next six weeks. However there are a couple of points that seem obvious and which should be emphasized.

1. The report is an excellent document. It presents results that are well thought out and which are appropriately qualified.

2. Some of the strong statements attributed to the document by the press are not found in the report. In particular, I have heard reports that the document suggests that dry deposition can be neglected in comparison to wet. This is not stated in the document although it does appear to be the personal opinion of at least one of the authors and was stated at a press conference concerning the document. In reality, of course, dry deposition is a nearer-source phenomenon than wet. If we are solely concerned about tall-stack emissions from power plants, then clearly wet deposition will dominate. However, if we are worried about natural emissions, automotive exhausts, or other surface sources, dry will dominate wet. The corollaries are important.

   (a) People who study what comes down in rain will find a major role of tall-stack emissions, because precipitation is where such emissions are focused.

   (b) Other workers who study concentrations in air near the surface or dry deposition of particles will find a major influence of more local sources, for the same reason.

   (c) Those who address the problem from the point of view of sulfur only, and power-plant stack controls to reduce sulfur emissions, will find little need to worry greatly about dry deposition far from sources, other than to provide background data for purposes of comparison.

3. The question of chemical nonlinearities is handled nicely, but I hope there are more qualifying statments than I detected on my perusal.

   (a) The extremes corresponding to strong nonlinearity and negligible nonlinearity can be handled in the models that presently exist. A simple sensitivity test would have been very worthwhile.

   (b) The question of chemical nonlinearity has been around for a long time, and has received a lot of thought. A major contribution to net effective nonlinearity (i.e. between stack emissions and deposition to ecosystems) may indeed be chemical, but we must also worry about meteorological nonlinearity. That is, the tendency for air from the southwest to be far more humid and thundery, hence more like an efficient chemical reactor than drier air masses associated with other wind vectors. In essence, I guess I object to the chemical focus of the non-linearity issue. I feel that it is more a meteorological matter, and I think it has been blown out of proportion as an issue.

4. The utility of models is discussed, but not in as sanguine a fashion as some reports have indicated. Again, several points are obvious.

   (a) No organized model can hope to reproduce the randomness of nature. Thus no model can hope to relate (except in average terms) the experience at any single receptor to any specific, distance source. The focus on very detailed, deterministic models is not to provide a definitive tool for proving source/receptor relations, but rather to study the factors which combine to cause events. A description of the average can be developed by running such models often, but this is like using a statistical mechanics model of molecular kinetic energy to determine the temperature of a fluid by integrating the energies of molecules, calculated individually. We have a better tool available -- a thermometer.

   (b) The use of statistical models is appropriate. In fact, we understand a lot about long-distance dispersion, on the average, and about its statistics. This knowledge can be applied with confidence, but only to the case of long averaging times and to relatively large areas.

   (c) Attempts to consider single-sources and single receptors are certainly poorly founded. Such questions are posed not by statisticians or meteorologists, who know better, but by politicians and industry representatives. The matter is somewhat like wanting to base a statistical average on a sample of one.

The report does not seem to contain any outrageous statements about the use (and abuse) of models, but again such statements have been credited to it. I fear that the cause might again be the press conference, or contact with some individual panel members. Certainly there is a need to recognize the specific utility of different kinds of models, Eulerian, Langrangian, Statistical, and Statistical Trajectory, and to recognize that the entire field of modeling cannot be criticized because of the failure of one kind of model to answer questions best suited to another.

To a considerable extent, this is where the US/Canada exercise fell down. The work was based on a random selection of appropriate and inappropriate models, whose results were then combined without apparent sensitivity to the fact that there was a problem.

I hope that these hip-shooting comments are of use. From my personal point of view, I can only regret that I did not have the opportunity to see the proofs of the material attributed to me in the report. It is said far better in the EPA/CARP document, it has a lot of typos, and some of the editing seems to have changed the tone in some places.

Battelle
Pacific Northwest Laboratories
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Telephone (509) 376-8639
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August 23, 1983

Dr. Bernard Manowitz
Chairman, Dept. of Applied Sciences
Brookhaven National Laboratory
Upton, NY 11973

Dear Bernie:

In your capacity as Chairman of the National Laboratory Consortium for Acid Deposition, you requested that we, as a NLC member Laboratory, review the National Academy of Sciences report, "Acid Deposition Atmospheric Processes in Eastern North America." Three PNL scientists, W.G.N. Slinn, M.T. Dana, and R.C. Easter, have carefully and thoroughly reviewed the report. The three were selected because of their long-standing involvement in research on precipitation scavenging processes, cloud physics and chemistry, and more specifically in recent years, the "acid rain" problem.

The findings of the three scientists regarding deficiencies of the report were uniform and unanimous. The attached summary comments represent their consolidated view.

The focus of their review was to identify the report's inadequacies and to document the unsatisfactory treatment of the nonlinearity issue. It should also be stated that the report's discussion of future research needs was good and that our scientists concur in the needs identified and the emphasis given.

Very truly yours,

C. E. Elderkin

C. E. Elderkin,
Associate Department Manager
Geosciences Research and
Engineering Department

cc: D. E. Olesen, PNL Laboratory Director
W. R. Wiley, PNL Director of Research
W. J. Bair, PNL Program Manager

PNL REVIEW OF NATIONAL ACADEMY OF SCIENCES REPORT
"ACID DEPOSITION ATMOSPHERIC PROCESSES IN EASTERN NORTH AMERICA"

The objectives of the NAS study project are stated in terms of answering the questions (p. 24): "If this goal [reducing H^+ deposition by 50%] were adopted, by how much would emissions have to be reduced? Conversely, by how much would deposition rates be reduced if there were specific reductions in emission rates?" The committee (p. 139): "...concludes that there is no evidence for a strong nonlinearity in the relationships between average emissions and depositions in eastern North America." In this context, linearity means proportionality of deposition to emissions. Although considerable qualifications are applied to this conclusion, we believe that the limited available evidence, as generally well covered by the report, does not support such a conclusion, or, that an opposite conclusion (nonlinearity) could be equally supported.

We will concentrate this review on the two major observations which the NAS study uses to support the above conclusion: 1) the apparent spatial uniformity of the sulfate/nitrate (S/N) ratios in eastern North America, and 2) the similarity in time trends between sulfur and nitrate emissions and the Hubbard Brook deposition data. In addition, we will comment on the report's treatment of chemical reaction mechanisms.

Sulfate/Nitrate Ratios

The committee found that the ratio of sulfate to nitrate in deposition was "nearly constant" at many locations in eastern North America and similar to state-by-state emission ratios. In Europe, a latitude (or distance from major source regions) dependence was found (Fig. 4.16 and Table 4.7), the ratio increasing with distance. The committee claims no such dependence for North America, but in fact if one constructs a table like Table 4.7 for North America, a similar trend can be identified. The ratio contour on Figure 4.17 for 1.2 at about 2 degrees latitude from the midwestern source region (where the ratio is 1) and the 1.6 contour at about 5 degrees compare favorably with the European results. The European data extend much farther (over about 15 degrees latitude), and the committee seems to have not noticed this.

The report's argument seems to be that: NO_x is readily converted to nitric acid which is scavenged efficiently, S/N ratios in deposition are equivalent to those in emissions; thus, SO_2 must be efficiently scavenged, implying nearly complete aqueous phase oxidation (no oxidant deficit). This logic is based on annual and regional averages (Figs. 4.17, 4.18 and Golomb's analysis, referred to on page 134-135), but considerable variations occur on shorter time scales. The MAP3S/PCN data of table 4.8 show a S/N ratio of 1.4± .1 for the warm months and .86± .19 for the cool months. Our own analysis of an expanded data set (MAP3S, 1981; MAP3S, 1983) for four inland northeast U.S. sites shows an even greater seasonal variation if one compares summer (Jun-Aug) to winter (Dec-Feb): 1.6± .1, and .53± .21, respectively. Inadequate discussion of the seasonal variation is presented in the report, especially in light of the at least implied lack of oxidant deficit.

Explanation of the apparent spatial uniformity of the ratio "...requires considerable specualtion." There is little hard evidence (as is admitted) to relate this uniformity (if true) to the linearity question. Furthermore, resemblance of the emissions to depositions (Figs. 4.17 and 4.18) may be purely fortuitous. Going to finer detail than annual or region averages, one finds (Table 4.9) significant contraindications of linearity. The concluded efficient scavenging of SO_2 also seems to be contradicted by the data of Figure 4.1 which shows a large area of quite high SO_2 air concentration. The question of whether or not there is adequate oxidant (e.g., H_2O_2) to consume this SO_2 is not fully discussed in the report.

In summary, on the basis of the included evidence and argument, the conclusion of an absence of nonlinearity does not seem any more justified than a conclusion to the contrary. The potential popularization of the committee's conclusion would thus be detrimental to a proper scientific (and regulatory) resolution of the acidic deposition question.

Hubbard Brook Data

It is unfortunate that the committee was forced to place so much weight on the data from a single deposition monitoring site. We agree that the Hubbard Brook data are the best of more than 5 years duration, but the site is located

outside the region of highest recent acidic deposition (Figs. 4.3 and 4.4), and a single site's data--however high in quality--cannot serve to characterize a region, particularly if the measurements involve strong dependence on meteorological variations. The time trend regressions (on p. 119, p. 133, and Figure 4.19) require much more discussion of the statistical significance and validity. In particular, the tests of the assumptions regarding independence and normality (p. 119) need to be more fully detailed. Extreme caution should be exercised in evaluating apparent trends in light of several important variabilities (MAP3S, 1982):

- source configuration and fluctuation
- atmospheric behavior (storm dynamics, precipitation features)
- changes in sampling practice and land use.

Long averaging times, e.g., years, will tend to smooth some of these variations, but long-term climatological trends may influence the record--even over 15 or 18 years. Any viable conclusions regarding cause-effect relationships and predictions of future behavior are particularly difficult to draw with confidence, especially when considering an isolated data set. Failure of the report to adequately treat the potential variables or provide a detailed statistical case weakens the significance of the conclusions regarding the Hubbard Brook data. In particular, the "similarity" of the emission and deposition trends in Figure 4.19 is not sufficiently quantified.

Chemical Processes

The discussion of chemical processes in Section 3 (p. 75-82) overemphasizes gas phase reaction mechanisms relative to the aqueous phase, in conflict with the well balanced approach in Appendix A. In particular, a detailed argument is made to show that the model of Rodhe, et al. may be improved with the result of de-emphasizing nonlinear effects of the gas phase reaction process (p. 75-81). Thence, only brief mention of the potential non-linear effects of aqueous phase mechanisms (e.g., through limitation of oxidant) is given (p. 81-82). It is admitted that "Existing analytical data for H_2O_2 in clouds do not allow an unambiguous conclusion to be reached today on the possible importance of these nonlinear effects." However, somewhat of a case for linearity of aqueous phase processes is made in the conclusions to the section: "measurements of oxidant

concentration in cloud water, although limited, suggest that concentrations may be sufficient in eastern North America for complete oxidation of SO_2..." The relative importance of gas versus aqueous-phase mechanisms is not sufficiently treated, and the conclusion regarding the linearity of the latter is ambiguous. Therefore, there is no firm support for the linearity argument from the study of chemical processes.

References

MAP3S (1980). The MAP3S Precipitation Chemistry Network: Fourth Periodic Summary Report, (1980), PNL-4144, Battelle, Pacific Northwest Laboratory, Richland, WA. Dec. 1981.

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GENERAL COMMENTS ON ACID RAIN
A Summary by the Acid Rain Peer Review Panel for the
Office of Science and Technology Policy
Executive Office of the President
June 27, 1983

The United States and Canada together are emitting annually more than 25,000,000 tons of sulfur dioxide (SO_2) and a comparable amount of nitrogen oxides (NO) and (NO_2), abbreviated as (NO_x), and these oxides can be converted by atmospheric chemical processes into sulphuric (H_2SO_4) and nitric (HNO_3) acids. The emissions are large enough to increase appreciably the acidity of natural rainfall, and rain in most of eastern Northern America is considerably more acid than that expected from natural processes alone. The Clean Air Act of 1970 marked the formal recognition by the U.S. government of the importance of reducing the emissions of sulfur to the atmosphere, and new power plants constructed since that time do control such emissions to lower levels. Such controls were a prudent first step. We recommend that additional steps should be taken now which will result in meaningful reductions in the emissions of sulfur compounds into the atmosphere beginning with those steps which are most cost effective in reducing total deposition.

The incomplete present scientific knowledge sometimes prevents the kinds of certainty which scientists would prefer, but there are many indicators which, taken collectively, lead us to our finding that the phenomena of acid disposition are real and constitute a problem for which solutions should be sought:

(1) The emissions of SO_2 and NO_x in eastern North America are at least ten times larger from human activities than from natural processes.

(2) A substantial fraction of such emissions are observed to return as sulfate (SO_4=) and nitrate (NO_3-) in rainfall; a probably comparable amount returns as "dry" deposition through surface interaction processes which are more difficult to monitor than the "wet" deposition in rain.

(3) In eastern North America the areas receiving the largest amounts of these acid rains are found within and downwind from the major source regions.

(4) The acidity of precipitation, some streams and some lakes in these major receptor regions are greater than the "natural" levels.

(5) Although some kinds of lakes have been acid throughout their known history, others located in principal receptor areas have become appreciably more acid during the past ten or twenty years.

(6) These changes in lake acidity have been accompanied by major changes in the biological activity within them, often including the disappearance of some species of fish.

(7) The largest of such aquatic effects have occurred in regions in which acidity is not "buffered" by the presence of alkaline minerals.

(8) Major areas of eastern North America have been identified whose geological composition is characterized by the absence of any important buffering capacity.

(9) Extensive evidence exists for increasing forest damage in eastern North America during the past few decades. Evidence of acid deposition as the primary cause for such harmful ecological effects on forests and other non-agricultural soils is, at present, much less compelling than that for aquatic damage.

The overall scientific understanding of the various aspects of acidic precipitation is quite incomplete at the present time, and will continue to have major uncertainties well into the future. Some of these gaps in our knowledge are permanent because the necessary measurements were not made ten, twenty, or fifty years ago before the potential future utility of such information was recognized. Other gaps exist because the needed scientific techniques have not yet been perfected or have not been adapted to the scale required for measurements covering much of the entire Western Hemisphere. Some of the important information will require at least ten or twenty years of additional data collection to take full cognizance of atmospheric variability and atmospheric cycles. Biological systems are extremely complex and variable. Response and recovery of many of these systems to external stress will require long-term (decades) detailed study for full evaluation. For these reasons, any current scientifically-derived recommendations must be based upon an imperfect, always increasing, body of pertinent data whose quality and completeness can be expected to improve for decades. Recommendations based upon imperfect data run the risk of being in error; recommendations for inaction pending collection of all of the desirable data entail even greater risk of damage.

The chemical processing of SO_2 and NO_x into acids in the atmosphere potentially involves a very large number of chemical

reactions, and the relative importance of these various reactions changes drastically with time and location, often in response to varyng meteorological conditions. Sulfur and nitrogen can be removed from the atmosphere in various chemical forms, and by both dry processes at the surface and wet processes in rainfall. Measurements of SO_4= and NO_3- in rainfall are now widespread, but do not have a long historical base. Measurements of dry deposition are so scattered (and experimentally doubtful) that quantitative assessment is essentially not possible even now.

The modeling of atmospheric emissions, transport and deposition has been confined almost entirely to the sulfur cycle, leaving nitrogen (and all else) to the future. The existing models do not agree with one another, and cannot be verified by comparison with observation because of the scarcity of good field data. They actually do not do very well in reproducing the observations on gaseous SO_2 that are available. Such models cannot be relied ukpon for (a) estimation of how much material emitted at A will be deposited at B; and (b) how much SO_2 will have been first converted to H_2SO_4.

There exists now no acceptable method for the determination of source/receptor relationships on a scale much smaller than "eastern North America." With a very large effort in laboratory atmospheric chemistry, in field measurements, and in atmospheric modeling, it might be possible within ten years (but certainly not five years) to produce a source/receptor model for eastern North America. We have great hope that methodology based on the use of natural tracers in fossil fuels may bypass some of these difficulties and perhaps reduce the time needed to elucidate this complex of problems. When a verified model exists in the future, there is a possibility that the source/receptor relationship will be sufficiently complex and variable that similar emission controls would still need to be assigned over rather large areas rather than locally.

Reduction below present SO_2 emission levels would reduce total sulfur deposition levels and as a consequence both reduce the probability for major changes in additional acid sensitive lakes or forests and allow the possibility for a return toward the original biological conditions existing in recently acidified areas.

The effects of acid deposition on biological systems in North America varies from certain to speculative. There is no question that some fresh water bodies have been altered

in sensitive areas. The increase of acidity can reach levels which result in the release or "mobilization" of aluminum from solid minerals raising the possibility of toxic metal effects on biological species in both lakes and forest soils. There is strong evidence for damaging effects on limestone monuments, bridges and buildings, and other structures, but there is no good estimate of the economic magnitude of these effects.

The effect of air pollutants on agriculture may be important but the quantitative evidence is scanty. (An estimate for ozone damage to agriculture in the United States is five percent of the cash value. We anticipate that the overall effect of acid precipitation on crops could be comparably significant.)

There is a tendency in the scientific literature to speak of "long-term" and "short-term" effects, or of "irreversible" and "reversible" changes. Damage to fresh water lakes, where it exists, may require a recovery time varying from a few years to tens of years when the stress is removed. This variation depends upon the availability in the environment of species for colonization, the extent to which trace element composition has been altered, and similar factors. The recovery time of a stressed sylvan environment is probably several decades or more in New England and Canadian latitudes. With both forests and lakes, the term "irreversible" might be used for a recovery time which stretches beyond a few decades.

We as a committee are especially concerned about possible deleterious effects of a sustained increase in the acidity of unmanaged soils. Its microorganism population is particularly sensitive to a change in acidity. But it is just this bottom part of the biological cycle that is responsible for the recycling of nitrogen and carbon in the food chain. The proper functioning of the denitrifying microbes is a fundamental requirement upon which the entire biosphere depends. The evidence that increased acidity is perturbing populations of microorganisms is scanty, but the prospect of such an occurrence is grave. It may take many years of accumulation of acidity, from wet or dry deposition, before measurable consequences would be observed. Such an effect is "long-term" or "irreversible." It may take at least that many years or longer for the soils to revert to their original condition. It is this possibility which provides us with the greatest concern.

Acid Rain Peer Review Panel General Comments

"Acid rain" or acid precipitation belongs to a socially very important class of problems that have the superficial aspects of being amenable to a permanent solution achieved by a straightforward sum of existing technological and legislative fixes. This is very deceptive. Rather, this class of problems is usually not permanently solved in a closed fashion, but is treated more commonly to accommodate a steady increase in knowledge and understanding, taking various actions that appear most effective and economical at any given time.

It is in the nature of the acid deposition problem, that actions have to be taken despite incomplete knowledge. We have earlier given estimates of how long it may take to understand the "wet" chemistry, or the biological response. Reasonably accurate models incorporating relevant meteorology, chemistry, minerology and biology take even longer. If we take the conservative point of view that we must wait until the scientific knowledge is definitive, the accumulated deposition and damaged environment may reach the point of "irreversibility."

We feel that the proper initial approach is to select particularly economically effective steps to begin to reduce our concerns in the light of gross transport and deposition features that have been identified, associated with seasonal and geographical variation. Purely as an example, it may be useful to consider having fuel of different sulfur content during different seasons since the efficiency for wet sulfuric acid deposition seems to be much less in winter. As other examples, first "least cost" steps might be gross reductions in sulfur emissions from non-ferrous smelters and intensifying coal washing.

DEPARTMENT OF ENVIRONMENTAL SCIENCES
CLARK HALL . UNIVERSITY OF VIRGINIA . CHARLOTTESVILLE, . VIRGINIA . 22903
(804) 924-7761

October 5, 1983

RECEIVED
OCT - 1983
COMMITTEE ON SCIENCE
AND TECHNOLOGY

Hon. Don Fuqua, Chairman
Committee on Science & Technology
U.S. House of Representatives
Washington, DC 20515

Re: Hearings on Acid Rain

Dear Mr. Chairman:

I wish to submit the enclosed material for the record in connection with recent hearings held by two of your sub committees.

My letter attempts to put in perspective the extent of economic damage that may be caused by acid deposition. In particular, my letter tries to rectify the impression that acid deposition damages agricultural crops and forests.

I am sending copies to other interested parties as shown.

Sincerely yours,

S. Fred Singer
Professor of Environmental Sciences
(Member of OSTP Acid Rain Peer Review Panel)

SFS:mrj

Enclosure

cc: Hon. James Sensenbrenner
Hon. James Scheuer
Hon. Raymond McGrath
Hon. John Dingell
Hon. James Broyhill

DEPARTMENT OF ENVIRONMENTAL SCIENCES
CLARK HALL . UNIVERSITY OF VIRGINIA . CHARLOTTESVILLE, . VIRGINIA . 22903
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September 30, 1983

Chairman
Committee on Science and Technology
U.S. House of Representatives
Washington, DC 20515

RE: Hearings on Acid Rain: Implications for Fossil Energy R&D

Dear Mr. Chairman:

I wish to comment on the Sept. 13 testimony read by Prof. W.C. Ackermann, Vice Chairman of the Acid Rain Peer Review Panel (established by the Office of Science and Technology Policy in the Office of the President).

The testimony was based on an interim report of the Panel, released on June 27, 1983 under the title "General Comments on Acid Rain." The report was drafted by a few members of the Panel but has not been fully discussed or agreed to by all of the Panel members. I have been a member of the Panel since its inception and I have reservations about items in the interim report dealing with the effects of acid rain. I have communicated these to the Panel Chairman and to OSTP and will discuss them in this letter.

In my view, acid rain is a serious problem but not life-threatening. It should be tackled without delay but cautiously.

I. Possible Crop Damage

In discussing damaging effects of acid rain on crops, the testimony, and interim report, imply the existence of large economic losses. I quote:

> **The effect of air pollutants on agriculture may be important but the quantitative evidence is scanty. (An estimate for ozone damage to agriculture in the United States is five percent of the cash value. We anticipate that the overall effect of acid precipitation on crops could be comparably significant.)**

I can not accept this implied assessment for the following reasons:

1. I have not seen support for the large numerical estimate for ozone damage, nor has it been critically reviewed by the Panel.

2. While ozone does indeed damage leaves, rubber tires, etc., ozone is not acid precipitation and has quite different action mechanisms (see below).

3. Therefore this extrapolation to acid precipitation is pure speculation, not supported by any data I know of, and certainly never reviewed by our Panel. (My research assistant has been unable to obtain any corroboration for acid rain damage from the Department of Agriculture).

4. In fact, our Panel has not seen any data, or even estimates, on the dollar amount of damage produced by acid deposition. Although we are sure that damage is produced, e.g. to certain lakes and to limestone structures, we could find no numerical values for such damage in all of the material which we were asked to peer-review.

II. Possible Soil Damage

The testimony, and the interim report of the Panel, express concern about increase in the acidity of unmanaged soils, and possible effects on micro-organisms, the food chain, and on forests. But: A) Evidence is contradictory. B) The argument contains a logical flaw which affects what should be done about emissions leading to acid rain.

A) Evidence for damage (or lack thereof)

I have searched for and will now cite recent authoritative reports, beyond the Working Group documents which our Panel was asked to review:

1. A summary of how soil acidification affects productivity was given by a group of distinguished academic researchers:

- soils amended in agricultural practice will not likely be negatively impacted by acidic deposition;
- soil acidification is a natural process in humid regions, so most soils that are easily acidified are already acid; and
- soils with low buffering capacity, relatively high pH, low sulfate adsorption capacity, no carbonates, and no basic inputs are susceptible to increased acidification rates from atmospheric inputs of acidic and acidifying substances. (1)

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(1) From: The Acidic Deposition Phenomenon and its Effects Vol. II. Chapter E-3: Effects on Vegetation, p. 3-1. EPA Office of Research and Development, Washington, DC 20460. EPA - 600/8-83-016B. May 1983.

That soil acidification is a natural process is also mentioned in a Science editorial (2) and is stressed in a recent survey article by two agricultural scientists. Their summary states:

> Acid rain is widely believed to be responsible for acidifying soil and water in areas of North America and northern Europe. However, factors commonly considered to make landscapes susceptible to acidification by acid rain are the same factors long known to strongly acidify soils through the natural processes of soil formation. Recovery from extreme and widespread careless land use has also occurred in regions undergoing acidification. There is evidence that acidification by acid rain is superimposed on long-term acidification induced by changes in land use and consequent vegetative succession. Thus, the interactions of acid rain, acid soil, and vegetation need to be carefully examined on a watershed basis in assessing benefits expected from proposed reductions in emissions of oxides of sulfur and nitrogen. (3)

They conclude:

> We believe that the effects of changing land use and vegetative succession cannot be dismissed on the basis of studies performed to date. Further, the hypothesis that increased deposition of acid and sulfate is causing equivalent leaching and acidification is theoretically unsound and is not supported by direct observations. Natural processes of acidification must be more carefully considered in assessing benefits expected from proposed reductions in emissions of oxides of sulfur and nitrogen. (3)

2. As far as effects on forest productivity are concerned, I quote the conclusions of the EPA study: such effects can be positive as well as negative.

- At present we have no direct evidence that acidic deposition currently limits forest growth in either North America or Europe, but we do have indications that tree growth reductions are occurring, principally in coniferous species that have been examined to date, that these reductions are rather widespread, and that they occur in regions where rainfall acidity is generally quite high, or pH is low (~ pH 4.3) for an annual average (Section 3.4.1).

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(2) P.H. Abelson, Rain Forests of Amazonia Science 221, p. 507, 5 Aug. 1983.

(3) E.C. Krug and C.R. Frink. Acid Rain on Acid Soil: A New Perspective. Science 221, pp. 520-524, 5 Aug. 1983.

- Controlled environment studies indicate that the deposition of acidic and acidifying substances may have stimulatory, detrimental, or no apparent effects on plant growth and development. Response depends upon species sensitivity, plant life cycle stage, and the nature of exposure to acidity. Some simulation studies have indicated that acidic deposition may result in simultaneous stimulation of growth and the occurrence of visible foliar injury (Section 3.4.1).

- The majority of crop species studies in field and controlled-environment experiments exhibited no effect on growth or yield as a result of exposure to simulated acidic precipitation (pH 3.0). In a few studies, though, growth and yield of certain crops were negatively affected by acidic deposition, while others exhibited positive responses (Section 3.4.2).

- A crop's net response to acidic deposition results from a combination of the positive effects of sulfur and nitrogen fertilization, the negative effects of acidity, and the interaction between these factors and other environmental conditions such as soil type and presence of other pollutants (Section 3.4.2).

- Available experimental results do not appear to indicate that the negative effects of acidic precipitation outweight the positive effects, however, many crops and agricultural systems have not been properly or adequately studied (Section 3.4.2). (4)

3. Much publicity has been generated recently about forest damage in Germany. During August 1983, I personally examined the documentation underlying the announcement of the German Minister of Interior, and held discussions with government scientists in Bonn and with forest scientists elsewhere in Germany.

(i) I do not find the evidence for a rapid "dying of forests" ("Waldsterben") credible or attributable to acid rain. For example, the German data shows that within 2 1/2 years the percentage of dead and sick trees increased from 15% to 90% (!) in certain areas.

(ii) If the data are indeed correct (i.e., not caused by a change in criteria), then other causes must surely be responsible, perhaps other air pollutants or even biological factors.

(iii) It is suspicious that the effects are seen in relatively non-industrial regions (Black Forest) but not in the Ruhr.

(iv) Incineration of wastes is widespread in Germany. The yearly emission of toxic heavy metals has been calculated as 600-800 tons. In addition, there is emission of chloro-organic chemicals (quantities unknown, but possibly herbicidal), as well as hydrochloric acid.

(4) Ref (1), p. 3-64.

4. The U.S. Department of Interior has attempted to separate the possible effects of acid rain from other air pollutants, such as photochemical oxidants (e.g., ozone and PAN), particulates (including trace metals), fluorides, sulfur dioxide and NOX. They conclude:

> b. Ecosystem Effects. The direct effects of acid deposition on forest vegetation in the field are generally unknown at the present time. In addition to the lack of evidence in support of direct acute injury, studies in North America and Norway utilizing tree ring analysis methodology were unable to detect reductions in growth attributable to acid deposition (Cogbill 1977; Abrahamsen et al. 1977; Strand 1980). (5)
>
> Therefore, unlike the other air pollutants discussed in this document, acid deposition appears to exert its primary influence on the soil component of forest ecosystems... (6)
>
> As for the acute and chronic injury results presented in subsection 2.4.2a, the significance of these effects on soil properties and processes in relation to soil fertility over the long term is presently unclear because of similar problems regarding the representativeness of the simulated "rain" solutions used (Wiklander 1979; Overrein et al. 1980; McFee and Cronan 1981) and the very large influence of site specific soil characteristics (such as soil mineralogy, texture, structure, permeability, pH, base saturation and organic matter, salt and sesquioxide contents) on the responses (Wiklander 1979; Bache 1980; Johnson 1980; Peterson 1980; Voigt 1980; Shriner 1981). In addition, counterbalancing processes (some of which are themselves initiated by acid deposition), such as increased weathering of primary minerals, deposition of nutrients (especially N and S) in incident precipitation, short term buffering by internal cycling of nutrients, and shifts in microbial community composition, may offset some of these responses (Abrahamsen and Dollard 1979; Aber et al. 1981; McFee and Cronan 1981). Finally, several researchers have noted the long term nature of a number of these anticipated responses, such as soil acidification, and have questioned the correctness of conclusions based on the one to two decades of observation available (Last et al. 1980; Linzon and Temple 1980). Current opinions of the future significance of these effects, based on present levels of acid deposition loading, are varied. They range from potential enforcement of latent nutrient deficiencies (other than nitrogen) induced by the fertilizing action of N added to the system in incident precipitation (Abrahamsen 1980)

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(5) The Effects of Air Pollution and Acid Rain on Fish, Wildlife and other Habitats. Report No. 6: Forests. Office of Research and Development, EPA, and Fish and Wildlife Science, Dept. of Interior, FWS/OBS-80/40.6, June 1982, p. 56.

(6) Ref. (5), p. 57.

to possible future reductions in forest productivity as a result of leaching losses of nutrient cations and mobilization of toxic concentrations of heavy metals (Abrahamsen and Dollard 1979; Overrein et al. 1980; Voigt 1980). (7)

B. "Irreversibility": A Logical Flaw

The Panel's interim report expresses concern that acidification of soils may be irreversible, i.e., a recovery span "beyond a few decades." But this concern is not consistent with our Panel's ultimate recommendation, i.e. that a "meaningful" reduction in emissions be made.

Any reduction in emission, and therefore presumably in deposition, cannot avert an "irreversible" fate but will only stretch out the time. For example, if certain soils were to be destroyed by 5 more years of acid rain (at the present level), then cutting the emissions by 50% (a major and extremely expensive undertaking) would only postpone the destruction by another 5 years.

If effects on soils were really important, then a radical cut in emission would be the only answer. For example, a 90% reduction would stretch the critical time to 50 years, which might be comparable with the recovery time.

I cannot recommend such a drastic course of action, however, based on what we know; nor do I believe would other Panel members.

Conclusion

The Interim report of our Panel, used as a basis for the Sept. 13 testimony, contains unwarranted extrapolations and speculations about economic and ecological damage of acid deposition.

I trust that our final Panel Report will reflect an agreed-to position of the Panel, including a closer evaluation of the terms "meaningful emissions reduction" and "least-cost approach" which appear in our overall recommendation.

Sincerely yours,

S. Fred Singer
Professor
(Member of OSTP Acid Rain Peer Review Panel)

SFS:lc

(7) Ref. (5), pp. 57-58.

First Annual Report on the Wisconsin Acid Deposition Research Program

Prepared by:

The Joint Acid Deposition Technical Review Committee

April 1983

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Julian Chazin - Air Management
Paul Koziar - Air Management
Jack Mason - Research
Robert Martini - Water Quality

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PREFACE

The Wisconsin Acid Deposition Research Program is a cooperative research effort among the Wisconsin Department of Natural Resources, the Wisconsin Utilities Association, the Wisconsin Public Service Commission, the Wisconsin Geological Survey, the University of Wisconsin, the U.S. Geological Survey, the U.S. Environmental Protection Agency, the U.S. Fish and Wildlife Service, and the Electric Power Research Institute.

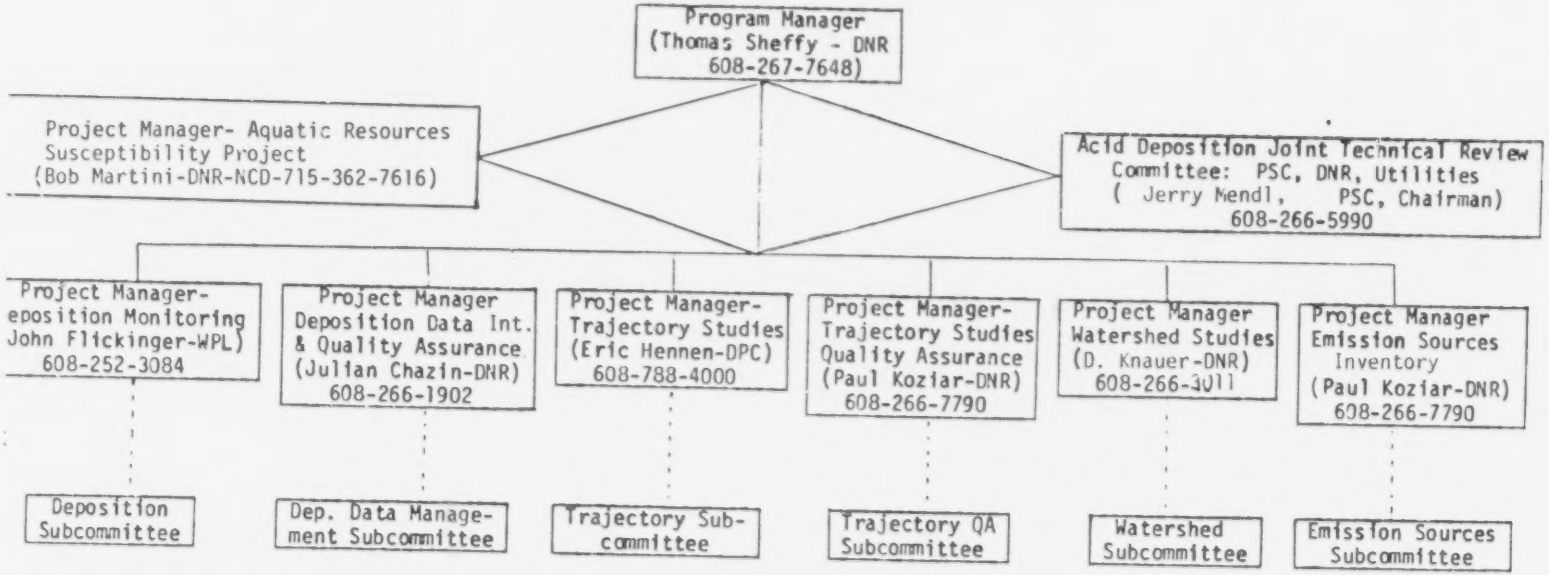
In the National Acid Precipitation Assessment Plan published by the U.S. Interagency Task Force on Acid Precipitation, the Wisconsin Acid Deposition Research Program was described as an "exemplary state program (which) reflects an integrated effort by state and private sector groups".

Following is a brief history of how this unique cooperative research program was developed.

The Wisconsin Department of Natural Resources first became involved in "acid rain" activities during the summer of 1979, when a general water quality survey of 350 lakes was conducted in north central Wisconsin. It was incidentally observed from this survey that approximately 40% of the sampled lakes had an alkalinity of 15 milligrams per liter (mg/l) or less. Based on information from Canada, softwater lakes with an alkalinity of 15 mg/l or less are considered susceptible to acid deposition. In the fall of 1979, rainfall samples were collected at several locations around Rhinelander. Analyses of the rainfall samples showed an average pH of 4.5. This preliminary information suggested a potential problem and prompted the formation of an Acid Deposition Task Force within the Department in February 1980. The Task Force examined all available information on acid deposition and published its findings in a report entitled "A Review of Acid Deposition in Wisconsin; Recommendations for Studying and Solving The Problem" in June 1980. The DNR presented its findings to the Wisconsin Public Service Commission and a joint Environmental Committee of the State Legislature in August 1980. It was agreed that further research was necessary. The Legislative Committee directed DNR to seek research funds from Wisconsin utilities, through the Public Service Commission. In turn, the PSC ordered Wisconsin utilities to develop and fund a research plan consistent with the objectives of a "Joint Acid Deposition Technical Review Committee" consisting of representatives from DNR, PSC, and the Wisconsin Utilities Association. The joint committee was formed in September 1980 and began developing a conceptual plan for acid deposition research in Wisconsin. By May 1981, technical work plans were developed by the joint committee for six research projects in the state. In October 1981, DNR also expanded its efforts in the North Central District into a seventh project with the help of federal funds. Taken together, these seven projects constitute the Wisconsin Acid Deposition Research Program (Figure 1 and Table 1).

This annual report describes the progress of these seven projects during the period of June 1, 1981 through December 31, 1982. A second annual report will be published on or about the same time in 1984 with a final report planned for early 1985.

Figure 1.
Wisconsin Acid Deposition Research Program
Table of Organization



- Project Managers will submit monthly progress/status reports to Program Manager.
- Program Manager will summarize program progress/status at quarterly meeting of Acid Deposition Joint Technical Review Committee.
- Program Manager will submit annual report of program status and findings to Acid Deposition Joint Technical Review Committee for approval.

Table 1. ORGANIZATION OF THE WISCONSIN ACID DEPOSITION RESEARCH PROGRAM

	Research Component	Objective	Methodology
1.	Deposition Monitoring Project	Characterize rainfall and dry deposition statewide.	Add 3 new precipitation monitors to existing network (8).
2.	Deposition Data Integration and Quality Assurance Project	Establish and maintain a reliable data bank.	Assemble historical precipitation data, implement quality assurance plan for existing monitors.
3.	Atmospheric Trajectory Study	Identify origin of air masses which deliver pollutants.	Backtrack 30 weather systems which deposit acidic precipitation at monitors.
4.	Trajectory Quality Assurance Project	Ensure correct interpretation of trajectory analyses.	Obtain independent review of trajectory plots.
5.	Emission Sources Inventory	Identify Wisconsin sources of SO_2 and NO_x; sources in surrounding states.	Assemble detailed emission inventory for Wisconsin and surrounding states.
6.	Intensive Watershed Study	Determine manner and rate of response of sensitive lake systems.	Measure interaction of acid deposition with vegetation, soil, groundwater, and lake in 2 representative watersheds.
7.	Aquatic Resources Susceptibility Project	Determine susceptibility, water quality changes; predict impacts.	Measure water quality and biota of a large set of lakes, compare to historical data, intensively study 2 representative lakes.

Executive Summary

The progress and preliminary findings of the seven research projects that constitute the Wisconsin Acid Deposition Research Program are reported for the period January 1, 1981 through December 31, 1982.

In the Deposition Monitoring Project, three new deposition monitors have been established at Round Lake (Douglas County), Legend Lake (Menominee County) and Lake Geneva (Walworth County). The data from these three new monitors and the existing eight monitors have been verified as part of the Deposition Data Integration and Quality Assurance Project, and show that Wisconsin is receiving acidic deposition with a mean pH of approximately 4.6.

The Atmospheric Trajectory Study and accompanying Trajectory Quality Assurance Project, intended to trace the origin of storm systems and polluted air masses which bring acid deposition to Wisconsin, are under way, but no data are available to date.

The Emission Sources Inventory has identified all major sources of SO_2 and NO_x in Wisconsin. Sources in surrounding states are being identified.

The Intensive Watershed Study at Round Lake (Douglas County) and East Eight Mile Lake (Bayfield County) is in its second year of data collection. No damage has been observed from acid deposition. The complete data set will be modelled next year to show when and under what conditions damage might occur.

The Aquatic Resources Susceptibility Project in north central Wisconsin has shown that about 30% of northern Wisconsin lakes are susceptible to acid deposition. However, a comparison of lake water quality data (pH, alkalinity, conductivity) from 1925-1941 to lake water quality data from 1979-81 has shown that no acid deposition damage has occurred to date. Numerous surveys of lake chemistry and aquatic life (plankton, macrophytes, fish) have been made for future comparisons.

A more detailed description and discussion of each project is provided in the body of this report. A summary of findings to date appears in Chapter 12.

1. Introduction

The popularized term "acid rain" actually refers to precipitation and dryfall (direct fallout of gases and acid particles) that contain unnaturally high amounts of sulfuric and/or nitric acid. Sulfuric and nitric acids are formed when sulfur dioxide (SO_2) and nitrogen oxide (NO_x) gases emitted by industrial, transportation and natural sources are chemically changed in the atmosphere. These gases can remain aloft for several days and can be transported hundreds of miles. The longer the time aloft, the greater the transformation of sulfur dioxide and nitrogen oxide into strong acids. The total amount of these strong acids reaching the ground by wet and dry deposition is often referred to as acid deposition.

The U.S. Environmental Protection Agency and the National Academy of Science estimate that in the U.S., over 90% of the sulfur dioxide in the atmosphere is emitted from man-made sources; 65% of this amount comes from coal-fired electrical generating plants. Automobiles emit about 50% of the nitrogen oxides, while utilities contribute another 30%.

Chemists characterize the acidity of precipitation by measuring its "pH". The pH of a solution is simply a numerical value on a scale that ranges from 0 (most acidic) to 14 (most alkaline). Pure water has a pH of 7.0 (the midpoint on the scale) and is exactly neutral. Values decreasing from 7.0 indicate increasing acidity. Values increasing from 7.0 indicate a solution is increasingly alkaline (Figure 1-1).

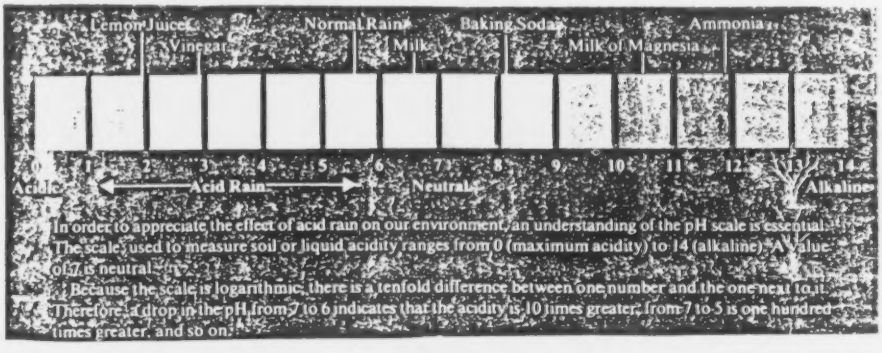

Lemon Juice
Vinegar
Normal Rain
Milk
Baking Soda
Milk of Magnesia
Ammonia
0
1
2
3
4
5
6
7
8
9
10
11
12
13
14
Acidic
Acid Rain
Neutral
Alkaline
In order to appreciate the effect of acid rain on our environment, an understanding of the pH scale is essential. The scale used to measure soil or liquid acidity ranges from 0 (maximum acidity) to 14 (alkaline). A value of 7 is neutral. Because the scale is logarithmic, there is a tenfold difference between one number and the one next to it. Therefore, a drop in the pH from 7 to 6 indicates that the acidity is 10 times greater; from 7 to 5 is one hundred times greater, and so on.

Figure 1-1. The pH Scale.

The average pH of rainfall over most of the Northeastern United States is about 4.3. Unpolluted rainfall is slightly acidic because some carbon dioxide from the atmosphere dissolves in rain, producing a weak solution of carbonic acid. Scientists sometime use a pH of 5.6 as a reference point for unpolluted rain because pure (distilled) water in equilibrium with atmospheric carbon dioxide has a pH of 5.6. The validity of this reference point is a subject of current debate. If a reference point of pH 5.6 is accepted, then present acid precipitation is approximately 1 pH unit lower (more acidic) than unpolluted rainfall. Because the pH scale is logarithmic, it is sometimes reported that acid deposition now occurring over much of the northeastern United States is approximately 10 times more acidic than normal.

The long term effects of acid deposition are documented in Sweden, in Norway, and in the Adirondack Mountains of New York. When the pH of lake water is lowered below 5.0 by prolonged acid loading, most fish cannot survive. Fish populations are particularly vulnerable in the spring. Acids stored in the snowpack can cause a rapid decrease in stream and lake pH when melting occurs. This phenomenon, called "spring shock", coincides with the spawning season for some species, jeopardizing fish reproduction. Other effects include the release of toxic metals (mercury, aluminum, zinc) from soil and lake sediment into lakes, with subsequent absorption by fish, loss of nutrients from soils, and degradation of man-made materials (buildings, roads, bridges, statuary).

The rate of acid loading is not the only factor to consider when estimating impact. Environmental degradation by acid deposition is also a function of geological sensitivity (susceptibility). Vast tracts of North America (Figure 1-2) consist of deep soils derived from limestone that are rich in carbonates which neutralize acid deposition. Lakes nestled in such soils (hard water lakes) have high alkalinity (acid-neutralizing capacity) and are not likely to be acidified.

The geology of northeastern North America is dramatically different. This region is typified by thin layers of soil, if any, which cover granite bedrock. Granite bedrock and overlying soils (tills) contain few, if any carbonates. Acid deposition can exhaust the limited amount of acid-neutralizing capacity contained in these soils. Likewise, the lakes located in such geologic regions are low in alkalinity (soft water lakes). Once this limited amount of acid-neutralizing capacity is used up, additional acid loading can depress the lake's pH. Unfortunately, much of the same region of North America that is susceptible also receives the most acid deposition (Figure 1-3). The "acid rain belt" where the pH of rainfall generally ranges between 4.1 - 4.7 extends from Minnesota to Nova Scotia, and from Florida to Hudson Bay.

Regions of North America Containing
Lakes Sensitive to Acidification
Source: After Galloway and Cowling, 1978 and United States Environmental Protection Agency, 1979

Figure 1-2. Acid-Sensitive Areas of North America.

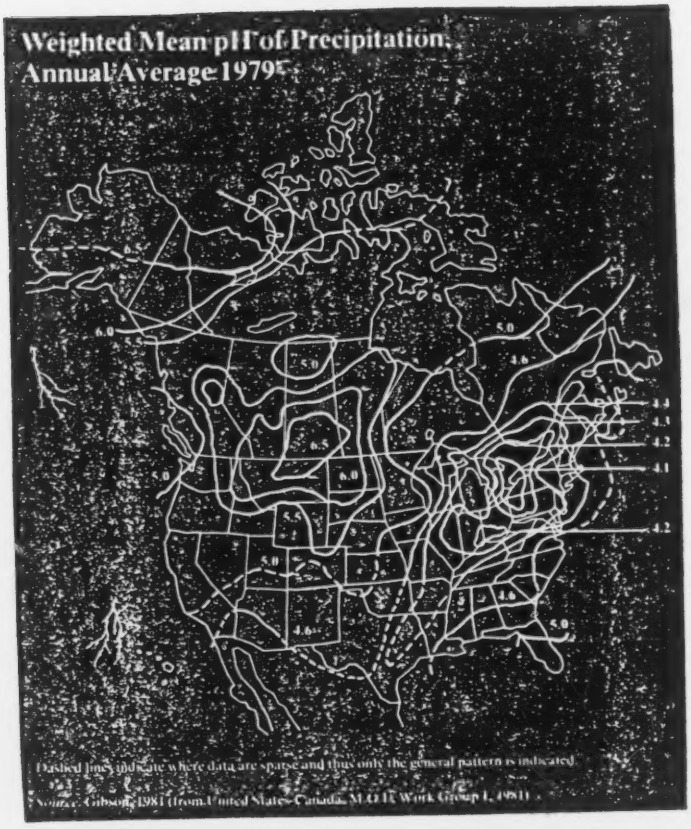


Figure 1-3. The Distribution of Acidic Precipitation in North America.

2. Key Questions to be Answered

In any research effort, it is critical to have agreement on the questions to be addressed. For the Wisconsin Acid Deposition Research Program, the salient questions that need to be answered are:

1) Is acid deposition occurring in Wisconsin?

   a) How much?
   b) Where?
   c) Under what circumstances?
   d) Is there or has there been a trend in acidity?

2) Where is the acid deposition coming from?

   a) How much of it is due to in-state air pollution sources?
   b) How much is due to long range transport from surrounding states or provinces?
   c) To what extent do Wisconsin's emissions affect other states and provinces?

3) Is acid deposition affecting Wisconsin's resources? If so, how much?

   a) Aquatic effects?
   b) Terrestrial effects?
   c) Economic effects?
   d) Effects on man-made materials?
   e) What are the anticipated future effects and rates of change which might be expected?

4) What and where are the significant sources of sulfur dioxide and nitrogen oxide?

   a) In Wisconsin?
   b) In surrounding states?
   c) In areas outside the U.S.?

5) What are the curtailment options?

However, the Wisconsin Acid Deposition Research Program, because of funding limitations, does not address the following:

1) Effects on Wisconsin farmland and crops.

2) Effects on man-made materials such as buildings, highways, bridges, culverts, and statuary.

3) Effects on mammals, birds, amphibians and reptiles.

4) Human health effects.

5) Mitigation strategies.

6) Potential economic impact.

7) Control strategies for sources, if necessary.

3. Objectives of Research Program

The objectives of the Wisconsin Acid Deposition Research Program are:

1) To quantify and characterize acid deposition throughout Wisconsin.

2) To identify the sources of acid deposition precursors and polluted air masses that cause acid deposition in Wisconsin.

3) a) To measure any effects of acid deposition on Wisconsin resources.
   b) To determine a rate at which resources might be affected if current deposition rates remain unchanged or increase.

4) To determine all sources contributing to acid deposition in Wisconsin.

5) To produce a credible report on the research, findings, and possible remedial actions.

4. Deposition Monitoring Project

Introduction

The Deposition Monitoring Project was established to meet three objectives:

1) Collect precipitation amount and chemistry data at three new sites in Wisconsin, to increase the data base for Wisconsin.

2) Collect dryfall amount and chemistry data at one of the precipitation sites, as part of an intensive watershed investigation.

3) Provide precipitation chemistry of events for use in trajectory analysis studies of storms that produced precipitation in Wisconsin.

The new network consists of three sites, one each located in northwestern, northeastern, and southeastern Wisconsin (Figure 4-1). These sites are managed by Battelle Pacific Northwest Laboratories, Richland, Washington. Data collection will proceed for 24 consecutive months from March 29, 1982, the date sample collection began.

Project Manager for the network is John Flickinger, Wisconsin Power and Light Company. Overall technical review is provided by the Deposition Monitoring Subcommittee of the Joint Acid Deposition Technical Review Committee with project responsibilities as shown in Figure 4-2.

Site Operations

Data collection began March 29, 1982, at the Round Lake and Lake Geneva sites. Due to contractural problems, operation of the Legend Lake site did not commence until April 23, 1982. Several improvements to the network as a whole were made during 1982 and are summarized below:

1) Original inking system on Belfort rain gauge replaced by a more readable felt tip system.

2) Two sensors were replaced because of slow recycle time.

3) Snow roofs were fabricated.

4) A revised site operators manual has been in use since September.

5) Greatly improved procedure's for marking Belfort charts have been in place since August.

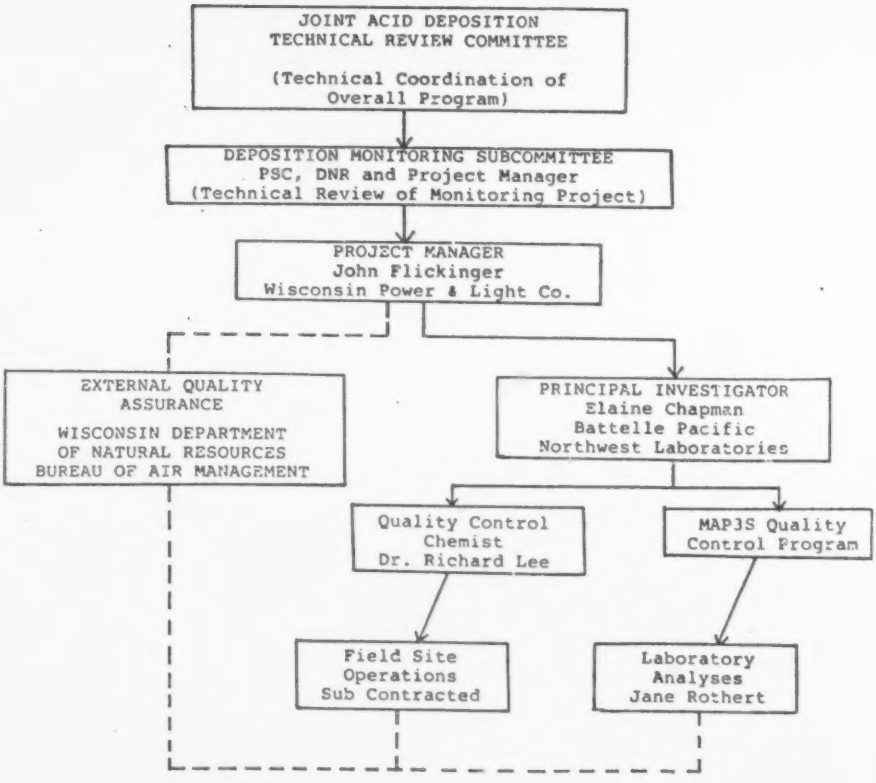
Operation summaries for each site are presented below, with descriptions of major problems encountered.

Round Lake - Sampling proceeded into May without the occurrence of serious problems. Data were lost in May, June, and July due to an injury to the principal site operator and his later failure to service the site on a regular basis. These actions resulted in the operator's termination and replacement late in August. Site operation under the new operator has been without incident since August.

Figure 4-1. Location of Wisconsin Acid Deposition Research Program Monitors.

28-916 0 - 84 - 65

Figure 4-2. Organization and Reporting Structure of the Deposition Monitoring Project



Instrument problems associated with the conductivity measurements have resulted in return of this unit to the manufacturer for repair. A replacement unit is in use until the repaired unit is back in service at the site.

Legend Lake - This site has operated continuously since its April 23 startup. Minor problems associated with recordkeeping and poor pH electrode performance have been corrected. Both the Aerochem Metrics collector and the pH/conductivity meter appear to be performing well.

Lake Geneva - This site has operated continuously since March 29. Minor problems have been experienced with exhaustion of field supplies due to the large number of precipitation events, and occasional sample shipping delays caused by the shipping service.

Instrument problems associated with the pH/conductivity meter caused its return to the manufacturer for repair in September. A loaner unit has been in use at the site since that time while the original unit undergoes repair.

Laboratory Operations

Laboratory analysis of all samples received by Battelle has proceeded smoothly. A minor delay occurred early in the project due to the sudden failure of an atomic absorption (AA) unit used for calcium and magnesium analysis. These samples were run by plasma emission spectroscopy initially, and then rerun by AA to double check results. No data loss occurred as a result. The strong acid screening analysis originally proposed by Battelle as an alternative to a full acidity titration is still being evaluated for its effectiveness in determining strong acid content. In the interim, a full titration to pH 9.5 is made, and the standard total acidity endpoint of 8.3 is used to determine strong acid concentration. This approach will continue until sufficient data demonstrate the usefulness of the screening procedure.

Total alkalinity is included in the ion balance, based on the assumption that it is all present as HCO_3^-. This assumption resulted from considerable technical discussion at Battelle during which a problem in the standard methods alkalinity formula was discovered. All alkalinity data from the project are expressed using this revised formula.

All samples with volume remaining are stored frozen for possible future reanalysis. These samples should be valuable in resolving questions relating to major ion concentrations, should any arise.

Quality Assurance

An internal program of quality control based on the analysis of check samples and duplicates has uncovered operational and instrument problems and allowed correction of these. In addition, Batelle's project officer visited each site in September as part of the routine six month inspection. The DNR field auditor also visited each network site (Round Lake was reaudited after the site operator was replaced) and provided detailed comments pertinent to site performance to the project manager. Complete copies of all audits and evaluations will be included with the appropriate quarterly report.

Reports

Preliminary data for the period March 29 to December 31, 1982 have been reviewed by the DNR and data tapes were sent to the Atmospheric Trajectory Study project officer (C.T. Main, Inc.). Hard copy reports of these quarters are available, and include all pertinent quality assurance activities and findings. Deposition data are discussed in Chapter 5.

5. Deposition Data Integration and Quality Assurance Project

Introduction

Until recently there has been no effort to collect and evaluate the quality of historical and ongoing results from deposition monitoring projects in Wisconsin. The Deposition Data Integration and Quality Assurance Project forms the basis of a Wisconsin-specific information collection and evaluation center. The three main objectives of the project are:

1) To establish and maintain a deposition data bank for Wisconsin.

2) To provide quality assurance for all deposition monitoring in Wisconsin.

3) To report all deposition data in the data bank on a regular basis.

This will be accomplished by collecting all prior and existing data from deposition networks that operate one or more sites in Wisconsin. These data are subject to a quality assurance evaluation, and entered into a comprehensive data base for deposition monitoring. To assure data quality of ongoing deposition monitoring, a program of field auditing, round robin testing, and data entry quality control has been established.

Project manager for this element of the research program is Julian Chazin, Chief, Air Monitoring Section, Wisconsin Department of Natural Resources. Technical review is provided by the Deposition Monitoring Subcommittee of the Joint Acid Deposition Technical Review Committee with project responsibilities as shown in Figure 5-1.

Data Integration

The precipitation chemistry monitoring sites in Wisconsin consist of 3 National Atmospheric Deposition Program (NADP) sites, 4 Great Lakes Atmospheric Deposition (GLAD) sites, 3 Wisconsin Acid Deposition Research Program (WADRP) sites, and 1 site operated by the Minnesota/Wisconsin Power Suppliers Group (M/WPSG) (Figure 5-2). Available acid deposition data from all but the M/WPSG site have been obtained and entered into a comprehensive data base. Appropriate quality assurance labels have been assigned to the deposition data entered in this data base. Precipitation chemistry data have been obtained from NADP for the period January 1980 to August 1982, from GLAD for the period January 1981 to October 1982, and from WADRP for the period March 1982 to December 1982. This data is published quarterly by the Data Integration and Quality Assurance Project in the Data Summary Report on Acid Deposition Monitoring in Wisconsin (Vol. 1, Nos. 1-3). Copies of this report can be obtained from the Bureau of Air Management, Wisconsin Department of Natural Resources.

Other data bases pertaining to acid deposition monitoring have been assessed and found to be adequate. Hence, there was no need to incorporate acid deposition data from other states in the upper midwest as was initially proposed.

Figure 5-1. Organization and Reporting Structure of the Deposition Data Integration and Quality Assurance Project

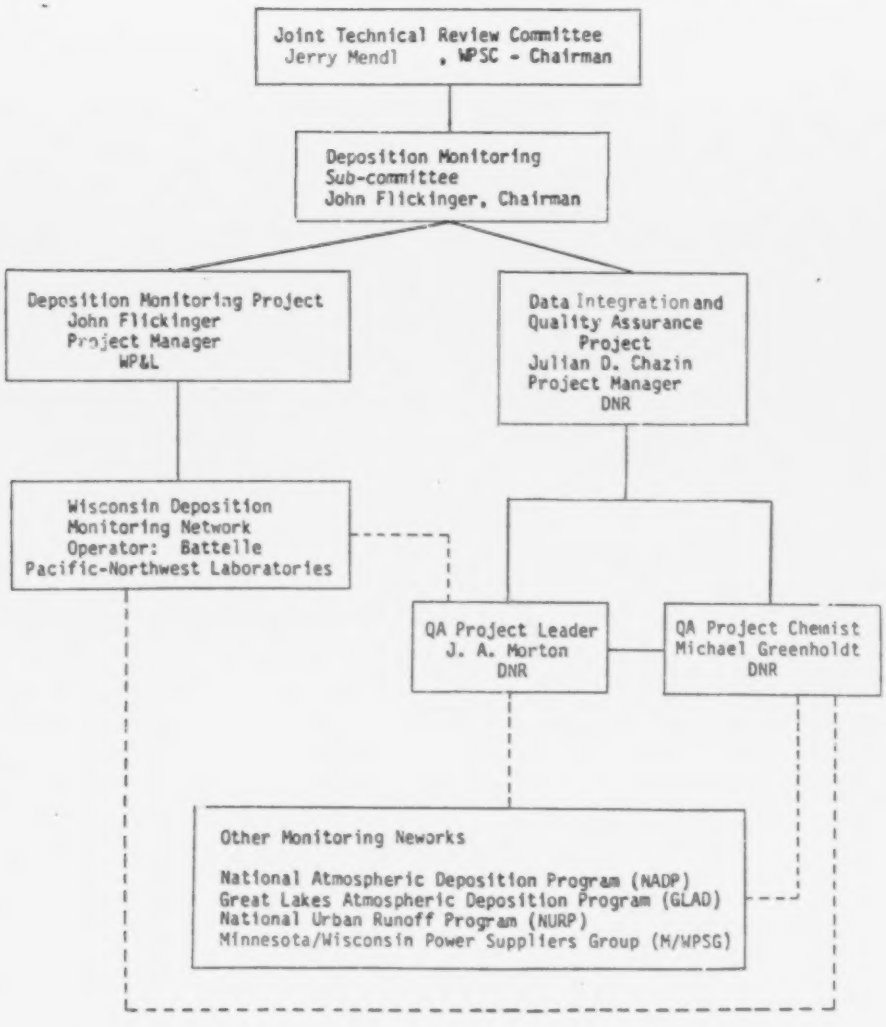


Figure 5-2. EXISTING WISCONSIN ACID DEPOSITION MONITORING SITES

MONITORING AGENCIES

- ■ NADP - National Atmospheric Deposition Program
- ● GLAD - Great Lakes Atmospheric Deposition Program
- ⊛ JTRC - Joint Technical Review Committee (Wisconsin PSC, Wisconsin Utilities, and Wisconsin DNR)
- □ M/WPSG - Minnesota/Wisconsin Power Suppliers Group

Quality Assurance

Quality assurance activities of the Data Integration and Quality Assurance project include conducting field audits of acid deposition monitoring sites, evaluating the central analytical laboratories of the monitoring networks, reporting on results of monitoring network participation in round robin and blind audit sample testing programs, validating data entered in the data base, and providing quality assurance assessments and information in the Data Summary Report on Acid Deposition Monitoring in Wisconsin (issued quarterly by the Data Integration and Quality Assurance Project).

According to the Quality Assurance Plan for Deposition Monitoring (Data Integration and Quality Assurance Project, July 1982) the following items have been accomplished:

1) Field audits of the NADP, GLAD and WADRP monitoring sites were conducted August-November 1982. Site selection evaluation, instrument audits, performance audits and equipment checks were included in the field audits.

2) The results of each monitoring site's participation in the USGS Intersite Comparison (Round Robin) Program are being received on a regular basis. These test results are used in the quality assurance assessment of each site.

3) The results of internal quality assurance and quality control activities are regularly obtained and are incorporated in the quality assurance assessment of each network's deposition monitoring data.

4) Before any precipitation data is incorporated into the data base, it has been screened to identify invalid values.

Comparison of Network Quality Assurance Activities

Shown in Table 5-1 is a comparison of the QA/QC activities of the NADP, GLAD, WADRP and M/WPSG monitoring networks. A more detailed description of the quality assurance pertaining to the M/WPSG network is described in the Precipitation Monitoring Quality Control Manual (Minnesota/Wisconsin Power Suppliers Group, 1981). Quality assurance activities for NADP are thoroughly described in the NADP Quality Assurance Report by Stensland et. al. (1980). The GLAD Quality Assurance procedures and tests are described in the Quality Assurance Project Plan: Policy, Procedures and Practice (DRAFT) (U.S. Environmental Protection Agency, Region V, 1982).

Data Reporting

The Data Integration and Quality Assurance Project has provided quarterly reports on the deposition monitoring (Data Summary Report on Acid Deposition Monitoring in Wisconsin). Copies of these reports can be obtained from the Bureau of Air Management, Wisconsin Department of Natural Resources. In addition, annual data reports will be issued.

Table 5-1. COMPARISON OF GLAD, NADP, N/WPSG and WADRP QA/QC ACTIVITIES

QA/QC Activity	GLAD	Network NADP	N/WPSG	WADRP
1. Blanks:				
A. Laboratory blank	X	X (Deionized water blank)	X	X
B. Reagent blank	X	X		
C. Field blank	X	X (Bucket blank)		
D. Filter blank		X		
2. Duplicates:				
A. Field duplicates	X		X	
B. Laboratory duplicates	X	X		For samples with suspect ion balance only
C. Standard duplicates		X		
D. Field check sample duplicates		X		X
E. Reference sample duplicates		X		X
3. Spikes:				
A. Surrogate spike	X			
B. Spiked sample	X		X	
4. Quality Control Checks:				
A. Lab control sample	X	X (EPA reference solutions, pH reference solutions, and conductance reference solutions)	X	X
B. Blind sample	X			
C. Check sample		X		
D. Check sample (field)				X
5. Control Standard	X	X	X	X
6. Control Charts	X	X	X	X
7. Log Books	X	X	X	X
8. External QA (LAB)				
A. Interlab comparison programs	USGS (bi-monthly	USGS (bi-monthly)		USGS (bi-monthly)
B. Lab performance audits	Monthly	USGS (bi-monthly)		
C. Lab-functional audits	Quarterly	Annually		
9. External QA (Field)				
A. Field monitoring site performance audits	Annually (WDNR)	Annually (WDNR)		Annually (WDNR)
B. Intersite comparison programs	USGS quarterly	USGS quarterly		USGS quarterly
C. Check samples		Weekly Check (4.30 pH)		Unknowns sent quarterly
10. Internal Performance and System Audits:				
A. System Review By Managers	X	X	X	X
B. In-House QC Audits	X	X	X	X
C. Control Limit Update	X	X	X	X
D. Methods Review	X	X	X	X

Network Descriptions

On the following pages are detailed descriptions of all deposition monitoring sites in Wisconsin.

Summary Laboratory pH Data for Precipitation Samples

All data and calculations discussed below are based on laboratory measurement of pH as opposed to field measurement of pH.

The arithmetic monthly mean pH, the geometric monthly mean pH, and the mass/volume weighted mean pH for precipitation samples collected at the WADRP and the NADP monitoring sites are presented in Table 5-2. Lab pH values of precipitation collected at the WADRP and the NADP monitoring sites are shown in Figures 5-3 and 5-4 respectively. The "arithmetic mean" represents the most common pH value of all the precipitation events measured. The "geometric mean" is the true average pH of all the precipitation events since it is based on the average hydrogen ion concentration.[1] This represents the true average acidity of the rainfall which has fallen over the time period (monthly). However, the impact of the precipitation acidity upon the environment is best assessed by the mass/volume-weighted mean pH. This is determined by weighting the pH of each event (expressed as the hydrogen ion concentration) by the amount of precipitation which falls during the event.[2]

For the WADRP sites, the arithmetic monthly mean pH ranged from 4.39 to 5.20, the geometric monthly mean pH ranged from 3.97 to 4.97, and the mass/volume weighted monthly mean pH ranged from 4.06 to 4.88 (Table 5-2). For the NADP sites, the arithmetic monthly mean pH ranged from 4.63 to 5.38, the geometric monthly mean pH ranged from 4.10 to 5.07 and the mass/volume weighted monthly mean pH ranged from 4.10 to 5.13 (Table 5-2).

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[1] This is accomplished by converting each pH value (the negative logarithm of the hydrogen ion concentration) to its respective hydrogen ion concentration, determining the arithmetic average of the hydrogen ion concentration per event and reconverting to the equivalent (logarithmic) pH value. This is the "geometric mean" pH.

[2] This is accomplished by converting the pH values for each event to their equivalent hydrogen ion concentration, multiplying by the respective precipitation amount for that event (whether expressed as volume or mass - for this purpose it doesn't matter). The product for all events is summed and the mass/volume average hydrogen ion concentration is obtained by dividing this sum by total amount of precipitation. This result is converted to the equivalent pH value to obtain the mass/volume weighted mean pH.

Description of Acid Depositon Monitoring Sites

Network:

Wisconsin Acid Deposition Research Project (WADRP)

Contact: John Flickinger
Wisconsin Power and Light Co.

Site Name: Lake Geneva

Site Address:

Clearing on north-central part of watershed in town of Lake Geneva, Walworth County, Wisconsin on Elk Club Road.

Site Operator:

Battelle-Pacific Northwest Laboratories

Station Environment: Rural

Latitude: 42°35' Longitude: 88°30'

Deposition Monitor:

Aerochem Metric Model 301

Sampling Frequency: Daily - Wet

Rain Gauges Present:

1. Belfort Recording Type
2. NWS

pH and Conductivity Meter:

Markson Model 4503 Selectromark Auto Analyzer (combined pH and conductivity meter)

Power: Line - 115 V AC

Network:

Wisconsin Acid Deposition Research Project (WADRP)

Contact: John Flickinger
Wisconsin Power and Light Co.

Site Name: Legend Lake

Site Address:

On County Road VV in field northwest of Legend Lake Lodge, in town of Keshena, Menominee County, Wisconsin.

Site Operator:

Battelle-Pacific Northwest Laboratories

Station Environment: Rural

Latitude: 44°53' Longitude: 88°43'

Deposition Monitor:

Aerochem Metric Model 301

Sampling Frequency: Daily - Wet

Rain Gauges Present:

1. Belfort Recording Type
2. NWS

pH and Conductivity Meter:

Markson Model 4503 Selectromark Auto Analyzer (combined ph and conductivity meter)

Power: Battery
(12-V deep cycle marine)

Network:

Wisconsin Acid Deposition Research Project (WADRP)

Contact: John Flickinger
Wisconsin Power and Light Co.

Site Name: Round Lake

Site Address:

Approximately 1 1/4 mile south of County Road Y in town of Wascott, Douglas County.

Site Operator:

Battelle-Pacific Northwest Laboratories

Station Environment: Remote

Latitude: 46°12' Longitude: 91°53'

Deposition Monitor:

Aerochem Metric Model 301

Sampling Frequency: Daily - Wet
Bi-Weekly - Dry

Rain Gauges Present:

1. Belfort Recording Type
2. NWS

pH and Conductivity Meter:

Markson Model 4503 Selectro Mark Analyzer (combined ph and conductivity meter)

Site Power:

Battery (12-V deep cycle marine batteries)

Network:

Great Lakes Atmospheric Deposition Program (GLAD) - Wisconsin

Contact: Bruce Rodger
Wisconsin Department of Natural Resources

Site Name: Milwaukee

Site Address:

On grounds of UW-Milwaukee north campus on roof of day care center at 2114 East Kenwood Avenue.

Site Operator:

Wisconsin Department of Natural Resources

Station Environment: Urban

Latitude: 43°05' Longitude: 87°53'

Deposition Monitor:

Aerochem Metric Model 301

Sampling Frequency: Weekly - Wet

Rain Gauges Present:

None present

pH Meter:

Digi-Sense 5994

Conductivity Meter:

Horizon Field Model C-1484-10

Power: Line - 115 V AC

Network:

Great Lakes Atmospheric Deposition Program (GLAD) - Wisconsin

Contact: Bruce Rodger
Wisconsin Department of Natural Resources

Site Name: Green Bay

Site Address:

Approximately 2 1/2 miles northeast of the city of Green Bay in open field at east end of UW-Green Bay campus.

Site Operator:

Wisconsin Department of Natural Resources

Station Environment: Rural

Latitude: 44°32' Longitude: 87°55'

Deposition Monitor:

Aerochem Metric Model 301

Sampling Frequency: Weekly - Wet

Rain Gauges Present:

None present

pH Meter:

Digi-Sense 5994

Conductivity Meter:

Horizon Field Model C-1484-10

Power: Line - 115 V AC

Network:

Great Lakes Atmospheric Deposition Program (GLAD) - Wisconsin

Contact: Bruce Rodger
Wisconsin Department of Natural Resources

Site Name: Manitowoc

Site Address:

Sampler located on elevated platform on roof of UW Center System campus building at 705 Viebahn Street in Manitowoc.

Site Operator:

Wisconsin Department of Natural Resources

Station Environment: Urban

Latitude: 44°05' Longitude: 87°39'

Deposition Monitor:

Aerochem Metric Model 301

Sampling Frequency: Weekly-Wet

Rain Gauges Present:

None present

pH Meter:

Corning Model 10

Conductivity Meter:

Horizon Field Model C-1484-10

Power: Line - 115 V AC

Network:

Great Lakes Atmospheric Deposition Program (GLAD) - Wisconsin

Contact: Bruce Rodger
Wisconsin Department of Natural Resources

Site Name: Cornucopia

Site Address:

West of Cornucopia in open field at top of cliffs at southwest corner of "Siskiwit Bay" located at northern end of Bell township in Bayfield County, Wisconsin, off Highway 13.

Site Operator:

Wisconsin Department of Natural Resources

Station Environment: Remote

Latitude: 46°52' Longitude: 91°08'

Deposition Monitor:

Aerochem Metric Model 301

Sampling Frequency: Weekly-Wet

Rain Gauges Present:

None present

pH Meter:

Digi-Sense 5994

Conductivity Meter:

Horizon Field Model C-1484-00

Power: Line - 115 V AC

Network:

National Atmospheric Deposition Program (NADP) - Wisconsin

Contact: Julian Chazin
Wisconsin Department of Natural Resources

Site Name: Spooner

Site Address:

On University of Wisconsin Agricultural Experiment Station 1 mile east of Spooner off of Highway 70.

Site Operator:

Wisconsin Department of Natural Resources

Station Environment: Rural

Latitude: 45°50' Longitude: 91°52'

Deposition Monitor:

Aerocnem Metric Model 301

Sampling Frequency: Weekly - Wet

Rain Gauges Present:

Belfort Recording Type

pH Meter:

Orion 601

Conductivity Meter:

Horizon Field Model C-1484-00

Power: Line - 115 V AC

Network:

National Atmospheric Deposition Program (NADP) - Wisconsin

Contact: Julian Chazin
Wisconsin Department of Natural Resources

Site Name: Trout Lake

Site Address:

Within the Wisconsin Department of Natural Resources Northern Highland State Forest 1/4 mile north of headquarters building off of County Road M.

Site Operator:

Wisconsin Department of Natural Resources

Station Environment: Remote

Latitude: 46°02'

Longitude: 89°38'

Deposition Monitor:

Aerochem Metric Model 301

Sampling Frequency: Weekly - Wet

Rain Gauges Present:

Belfort Recording Type

pH Meter:

Orion 407A

Conductivity Meter:

Horizon Ecology Field Model C-1484-00

Power: Line - 115 V AC

Network:

National Atmospheric Deposition Program (NADP) - Wisconsin

Contact: James Weinbauer
Consolidated Papers, Inc.

Site Name: Lake Dubay

Site Address:

East of Highway 51, near intersection of County Road XX and Highway 51, on Lake Dubay Dam, Portage County.

Site Operator:

Consolidated Papers, Inc.

Station Environment: Remote

Latitude: 44°39'52"

Longitude: 89°39'22"

Deposition Monitor:

Aerochem Metric Model 301

Sampling Frequency: Weekly - Wet

Rain Gauges Present:

Belfort Recording Type

pH Meter:

Orion 701A

Conductivity Meter:

YSI 32

Power: Line - 115 V AC

Network:

Minnesota/Wisconsin Power
Suppliers Group (M/WPSG)

Contact: Steven Lindenburg
Minnesota/Wisconsin Power
Suppliers Group (M/WPSG)

Site Name: Alma, WI

Site Address:

Off County Road N, 5.5 kilometers
southeast of the city of Alma,
Wisconsin in Buffalo County

Site Operator:

Dairyland Power Cooperative

Station Environment: Rural

Latitude: 44°30' Longitude: 44°30'

Deposition Monitor:

1. Refrigerated Automatic
Sequential Precipitation
Collector (Saeger Krupa
Sampler)

2. Anderson Model 3000
Dichotomous Air Sampler

Sampling Frequency:

Wet - Sequential/Subevent

Rain Gauges Present:

Tipping rain gauge located
1/2 mile away

pH Meter:

None present

Conductivity Meter:

None present

Power: Line

Table 5-2. Monthly Arithmetic Mean Lab pH, Monthly Geometric Mean Lab pH, and Monthly Mass/Volume Weighted Mean Lab pH of Wet Deposition Collected at Wisconsin Acid Deposition Research Program (WADRP) and National Atmospheric Deposition Program (NADP) Sites for 1982

		Months (1982)								
	Site	Jan.	Feb.	March	April	May	June	July	Aug.	Sep.
Arithmetic Mean Lab pH[6]	Round Lake[2]									4.72
	Legend Lake[2]					4.52	5.20	4.46	4.55	
	Lake Geneva[2]				4.45	4.59	5.20	4.45	4.39	
	Spooner[3]	4.80	5.20	5.16	5.22	5.38				
	Trout Lake[3]	4.68	4.68	4.63	5.14	5.38	4.66			
	Lake Dubay[3]							4.68	4.93	
Geometric Mean Lab pH[7]	Round Lake[2]									4.62
	Legend Lake[2]					4.24	4.97	4.28	4.47	
	Lake Geneva[2]				4.38	4.26	4.85	4.17	3.97	
	Spooner[3]	4.78	4.72	4.84	5.07	4.92				
	Trout Lake[3]	4.62	4.68	4.48	4.78	4.80	4.10			
	Lake Dubay[3]							4.65	4.57	
Mass/Volume Weighted Mean Lab pH	Round Lake[2,4]								4.80[1]	4.81
	Legend Lake[2,4]					4.06	4.88	4.47	4.48	
	Lake Geneva[2,4]				4.70	4.32	4.56	4.31	4.51	
	Spooner[3,5]	4.72	4.78	4.71	5.13	4.81				
	Trout Lake[3,5]	4.53	4.68[1]	4.56	4.69	4.55	4.10			
	Lake Dubay[3,5]							4.57	5.04	

	Site	Jan.	Feb.	March	April	May	June	July	Aug.
Total Precipitation Collected Per Month	Round Lake[9]								
	Legend Lake (grams)[2]					2,815	4,300	4,635	5,568
	Lake Geneva (grams)[2]				4,828	5,686	4,811	11,011	5,880
	Spooner (ml)[3,8]	1,496	245	1,492	2,608	6,469			
	Trout Lake (ml)[3,8]	1,473	521	1,379	8,377	7,144			
	Lake Dubay (ml)[3,8]							4,242	

[1]pH value represents one sample instead of a montnly mean.

[2]Event sampling

[3]Weekly sampling; data preliminery - not fully quality assured by NADP

[4]Mass (grams) used in weighted mean pH calculation

[5]Volume (ml) used in weighted mean pH calculation

[6]Most common pH

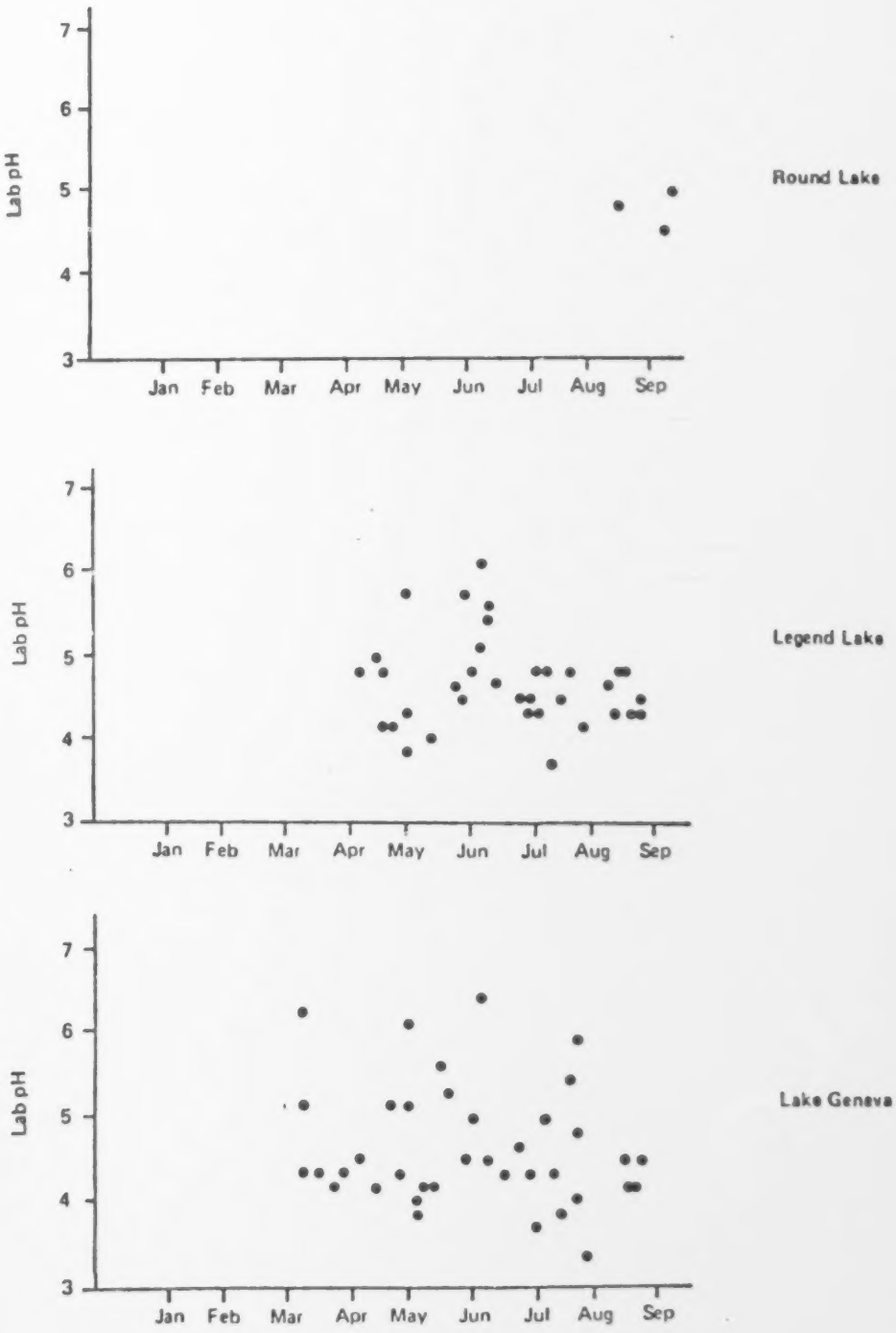
[7]True Mean

[8]Trace precipitation samples and samples diluted for analysis were not included in totals of precipitation collected. Trace and diluted samples did not constitute over 1% of the total precipitation collected for the sampling period at each site.

[9]Entire month of precipitation was not collected at the Round Lake site; total precipitation collected not shown.

2514P-4

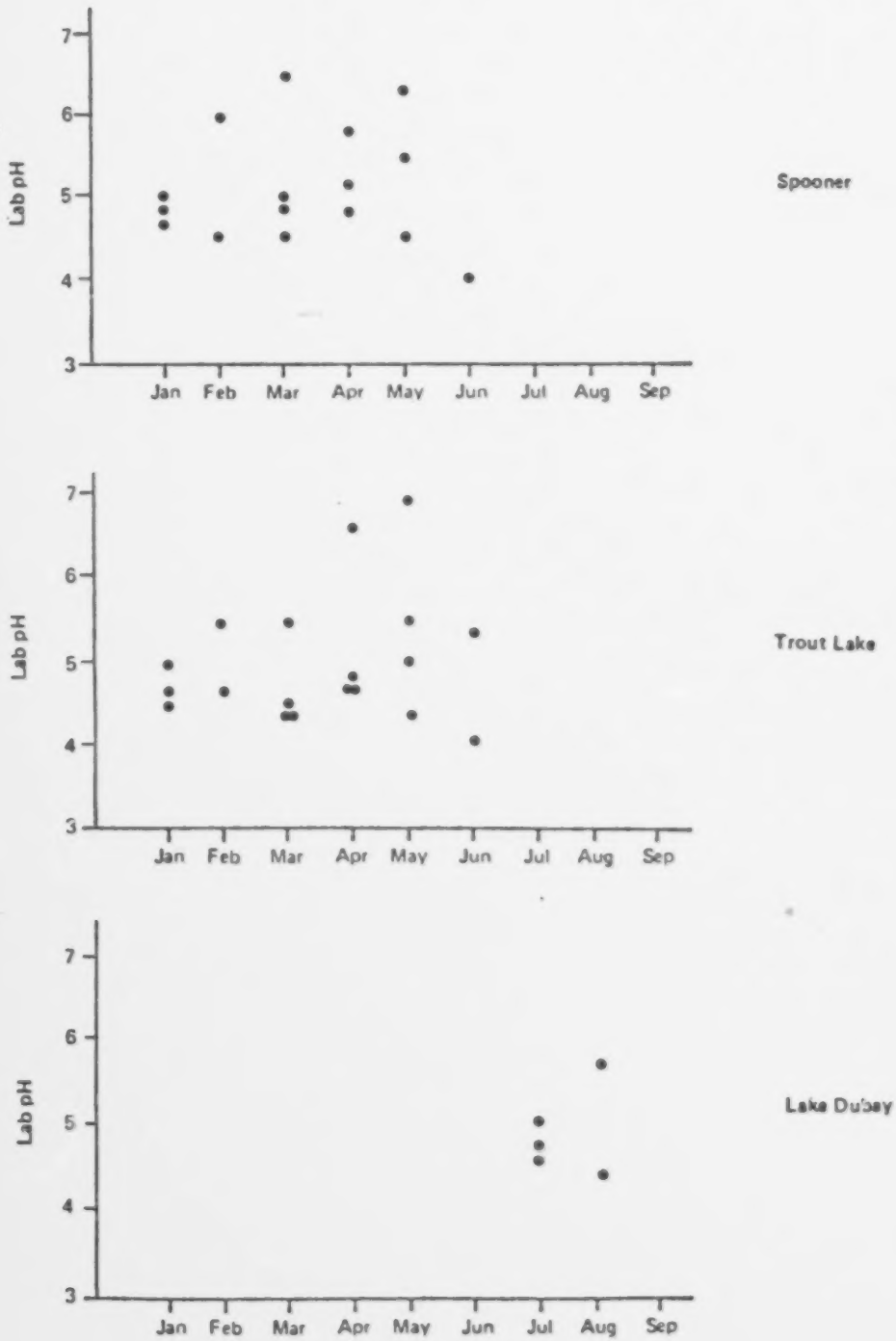
Figure 5-3. Lab pH Values of Wet Deposition Samples at the Round Lake, Legend Lake and Lake Geneva Monitoring Sites for 1982 (WADRP) (Event Sampling)



28-916 0 - 84 - 66

Figure 5-4. Lab pH Values of Wet Deposition Samples at the Spooner, Trout Lake and Lake Dubay Monitoring Sites for 1982 (NADP)

(Weekly Sampling)



Summary of SO_4 and NO_3 Concentrations of Precipitation Samples

Mean (arithmetic) monthly SO_4 and NO_3 concentration values of precipitation chemistry samples collected at WADRP and NADP monitoring sites are presented in Table 5-3. The mean monthly SO_4 concentration of precipitation collected at the WADRP sites for 1982 ranged from 1.18 mg/l to 4.48 mg/l. For the NADP sites, the mean monthly SO_4 concentration ranged from .67 mg/l to 5.74 mg/l for 1982. The mean monthly NO_3 concentration for deposition collected at the WADRP sites ranged from 1.02 mg/l to 3.59 mg/l. The mean monthly NO_3 concentration of precipitation collected at the NADP sites ranged from 1.32 mg/l to 4.14 mg/l for 1982. As shown in Table 5-3, the SO_4 and NO_3 concentrations of single samples collected at the NADP sites were higher than the values cited above, but they do not represent a monthly mean.

Total Monthly SO_4 and NO_3 Deposition Loading (Kg/Ha)

The sulfate (SO_4) and nitrate (NO_3) total monthly deposition loading (Kg/Ha) for precipitation collected at the WADRP and NADP monitoring sites are presented in Table 5-4. The calculated deposition loading provides the amount (mass) of a substance that is deposited on a specified land area. The SO_4 and NO_3 loadings were calculated from the SO_4 and NO_3 concentrations (mg/l) in the following manner. First, the actual weight of SO_4 and NO_3 collected as part of each precipitation sample is determined. This provides us with the weight of SO_4 and NO_3 that is deposited on an area the size of the precipitation collector bucket (.068 m^2). This gives us the SO_4 and NO_3 deposition amount which is expressed in milligrams/meter^2 (mg/m^2). For the final deposition loading value, the deposition amount in mg/m^2 was converted to Kilograms/Hectare (Kg/Ha).

For the WADRP monitoring sites, the total SO_4 loading per month ranged from 1.14 Kg/Ha to 4.24 Kg/Ha. The total monthly NO_3 loading ranged from .65 Kg/Ha to 2.24 Kg/Ha for the WADRP monitoring sites. The total monthly SO_4 deposition loading for the NADP monitoring sites ranged from .05 Kg/Ha to 3.05 Kg/Ha. The total monthly NO_3 deposition loading for the NADP monitoring sites ranged from .07 Kg/Ha to 1.91 Kg/Ha.

Table 5-3. Mean (Arithmetic) Monthly Concentration of SO_4 and NO_3 of Wet Deposition Collected at Wisconsin Acid Deposition Research Program (WADRP) and National Atmospheric Deposition Program (NADP) Monitoring Sites for 1982

		Months (1982)								
	Site	Jan.	Feb.	March	April	May	June	July	Aug.	Sep.
SO_4 Mean Monthly Concentration (mg/l)	Round Lake[1]								1.34[3,4]	1.18[4]
	Legend Lake[1]				3.36[3,4]	4.51	1.73	2.13	2.01	1.19[4]
	Lake Geneva[1]			2.98[3,4]	4.48	3.72	2.94	4.40	3.09	
	Spooner[2]	1.40	1.23	2.61	2.30	2.05	10.26[3]			
	Trout Lake[2]	.67	.91[3]	2.18	3.47	5.74	5.89[3]			
	Lake Dubay[2]							2.67	4.02[4]	
NO_3 Mean Monthly Concentration (mg/l)	Round Lake[1]								2.91[4]	1.27[4]
	Legend Lake[1]				2.48[3,4]	3.59	1.02	1.78	1.24	1.30[4]
	Lake Geneva[1]			1.12[3,4]	2.85	2.65	2.06	2.85	2.45	
	Spooner[2]	1.54	2.02	1.45	1.75	1.32	2.43[3]			
	Trout Lake[2]	1.67	1.59[3]	1.55	2.21	4.14	2.35[3]			
	Lake Dubay[2]							2.20	1.95[4]	

[1]Mean concentration per event (event sampling)

[2]Mean concentration per week (weekly sampling); data preliminary - not fully quality assured by NADP

[3]Value represents one sample

[4]Sampling period does not include entire month.

2514P-5

Table 5-4. Total Monthly SO_4 and NO_3 Deposition (Kg/Ha) (Wet Precipitation) Collected at Wisconsin Acid Deposition Research Program (WADRP) and National Atmospheric Deposition Program (NADP) Sites for 1982.[1]

	Site	Months (1982) Jan.	Feb.	March	April	May	June	July	Aug.
SO_4 - Total Deposition Per Month (Kg/Ha)	Legend Lake[2]					2.06	1.14	1.36	1.53
	Lake Geneva[2]				2.06	2.63	2.35	4.24	1.86
	Spooner[3,4]	.20	.05	.54	.89	2.03			
	Trout Lake[3,4]	.17	.07	.49	2.48	3.05			
	Lake Dubay[3,4]							1.57	
NO_3 Total Deposition Per Month (Kg/Ha)	Legend Lake[2]					1.76	.65	1.03	.84
	Lake Geneva[2]				1.14	1.75	1.25	2.24	1.34
	Spooner[3,4]	.32	.07	.30	.68	1.28			
	Trout Lake[3,4]	.43	.12	.32	1.41	1.91			
	Lake Dubay[3,4]							1.20	
Total Precipitation Collected Per Month	Legend Lake (grams)[2]					2,815	4,300	4,635	5,568
	Lake Geneva (grams)[2]				4,828	5,686	4,811	11,011	5,880
	Spooner (ml)[3,5]	1,496	247	1,492	2,608	6,469			
	Trout Lake (ml)[3,5]	1,473	521	1,376	8,377	7,144			
	Lake Dubay (ml)[3,5]							4,242	

[1]The Round Lake monitoring site (WADRP) was not included in this table because precipitation sampling data is not available for an entire month.

[2]Event sampling

[3]Weekly sampling; data preliminary - not fully quality assured by NADP

[4]Trace precipitation samples and samples diluted for analysis were not included in these total deposition calculations. Trace and diluted samples did not constitute over 1% of the total precipitation collected for the sampling period at each site.

[5]Trace precipitation samples and samples diluted for analysis were not included in totals of precipitation collected. Trace and diluted samples did not constitute over 1% of the total precipitation collected for the sampling period at each site.

2514P-6

6. Atmospheric Trajectory Study

Introduction

The primary objective of the Atmospheric Trajectory Study is to identify the source regions of acid precipitation precursors that were found to have contributed to acid deposition in the State of Wisconsin. In addition, in-state versus out-of-state contributions will be identified to the extent possible. The method chosen to accomplish this is the reverse trajectory method.

Air mass trajectories will be calculated for 30 precipitation events. Using this case study approach, the trajectories, meteorology and precipitation chemistry for each of these 30 events will be evaluated. In addition, a climatological approach will be used to classify and evaluate the overall meteorological and chemical data base.

It is hoped that by evaluating in a general sense, all of the data through the climatological approach and in a specific sense, the 30 case studies in detail, increased knowledge can be developed of the chemical and physical processes of the acid deposition phenomenon as well as a greater understanding of the source regions of acid deposition in the State of Wisconsin.

Project Manager for this study is Eric Hennen, Dairyland Power. Technical review is provided by the Atmospheric Trajectory Subcommittee of the Joint Acid Deposition Technical Review Committee with project responsibilities as shown in Figure 6-1.

Project Background

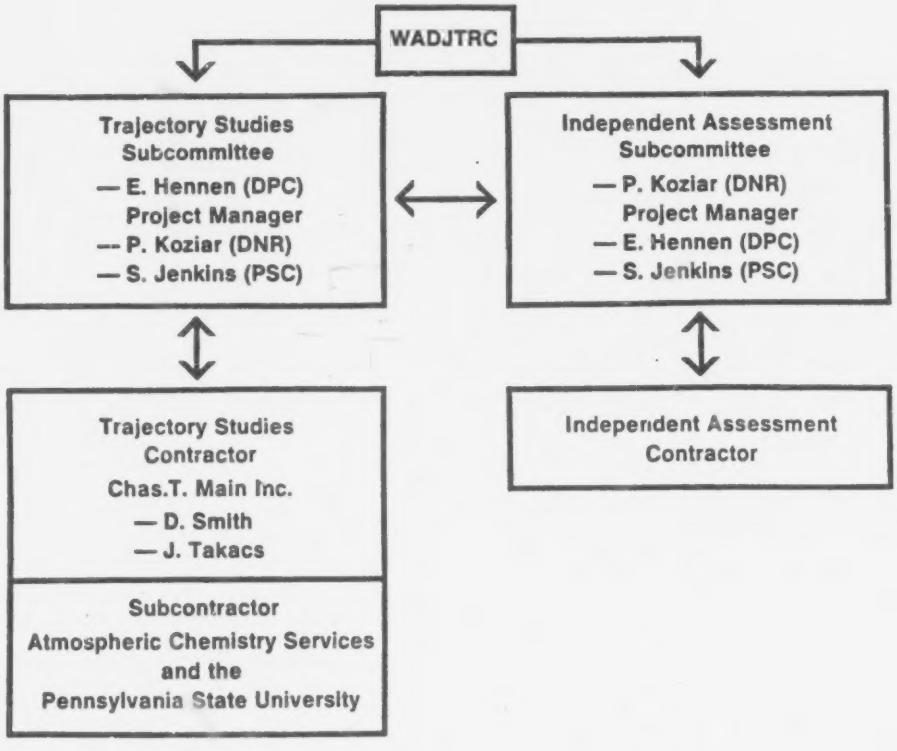
Early in the formative stages of Wisconsin's acid deposition study plan, a method for study of the atmosphere's role was needed. Two techniques exist: 1) long range transport modeling, and 2) reverse trajectory analysis.

Long-range transport modeling is the favored approach by some authorities as it allows for "quantitative" results, (even though the errors associated with these models can be extremely large). In the use of long range transport modeling, many assumptions have to be made:

1) The dispersion characteristics of the atmosphere. Usually the models assume only horizontal motion (no vertical motion). Interestingly enough, if only horizontal motion prevailed, we would have no rain.

2) The chemical transformations in the atmosphere. Usually SO_2 is assumed to react linearly to sulfate, with no dependence on other atmospheric chemical species. But in the preface to the long range transport modeling group (U.S. - Canadian Memorandum of Intent) report, the following is stated:

Figure 6-1. Organization and Reporting Structure of the Atmospheric Trajectory Study.



> "To date, these (long range transport) models have been successful in describing sulfate deposition on an annual basis. Hydrogen and nitrate ion deposition, two important factors in acid rain, have not yet been successfully incorporated in the models."

NOTE: "Successful" means within a factor of two.

It is clear that acid deposition is a function of the hydrogen ion concentration and not the sulfate ion. Yet, these models assume that sulfate is a surrogate for acid deposition by assuming a one to one correlation of hydrogen with sulfate ion. While this assumption may be good in the northeastern United States where the correlation between the sulfate and hydrogen is acknowledged to be high, deposition data in Wisconsin and Minnesota may not exhibit this high correlation.

It is clear that the use of these long range transport models requires an assumption in the underlying source/receptor relationship: That SO_2 reacts to form SO_4, which results in acid rain. However, there has been and continues to be much controversy surrounding this basic theory. Is the atmospheric process of acid deposition as simple as SO_2 to SO_4 to acid deposition or is more complicated? This is a very difficult question. Yet, surprisingly there are very few research programs underway to try to study and understand this basic source/receptor relationship. It was decided that the relationship between source emissions and acid deposition was not established in Wisconsin. Therefore, applying "off the shelf" long range transport models that inherently had an assumed relationship in their algorithm was not the preferred way to proceed. In order to establish this source/receptor relationship, a research effort incorporating a climatological analysis and reverse trajectory approach could establish this link. Thus, the reverse trajectory method was chosen.

The trajectory method does not treat chemical transformation at all. Instead, this approach takes a measured acid deposition event (from the deposition network described in Chapter 4) and works backward through the measured National Weather Service meteorology to find out from where the air feeding the rain producing storm came. By knowing the path of that air, it will be known what source areas could have contributed to the rain event, and which could not have. Because the trajectory methods does not consider atmospheric chemistry, it is not subject to the errors associated with these chemical assumptions. The trajectory method, in fact, is subject to only the atmospheric dispersion assumption used in long range transport models, and the trajectory method improves significantly on those assumptions. Refinements to the technique are discussed below.

Trajectory research is in its relative infancy. While trajectory methods have been employed since about 1940, it is only within the last five years that trajectory techniques have begun to be recognized as viable meteorological analytical techniques for determining the history of air parcels.

The most common trajectory method employed today involves the use of the AkL-ATAD model developed by J. L. Heffter of the National Oceanic and Atmospheric Administration. The model calculates backward air parcel motions on a constant level surface. Thus, no vertical motion is included (or allowed). This is a very common assumption in meteorology and, in general, is not a bad one. However, in order to have rain, you need clouds and vertical motion. Therefore, it is clear that in order to actually track a rain producing event, vertical motion must have occurred. The trajectory method selected must then incorporate this vertical motion to be accurate.

Atmospheric motion tends to be largely horizontal, following physical laws such as conservation of energy and momentum. Atmospheric motion also tends to be adiabatic[a] after the first law of thermodynamics. In the atmosphere, a parcel following adiabatic motion is also said to be isentropic[b].

In meteorology, adiabatic motion is also a good assumption and while air tends to blow largely horizontallyit is generally blowing along isentropic surfaces, which have some slight slope to them. Thus, air moving along these isentrophic surfaces actually has some small vertical component. Therefore, trajectories computed on an isentropic surface more correctly approximate the actual parcel path because vertical motion is implicit to the method. For this reason, the isentropic trajectory method was chosen for this project.

Project Description

The project officially started on March 29, 1982 with the start of meteorological data collection and coincident with the start of the Deposition Monitoring Project. The consultant contracted for the work is the Charles T. Main firm of Boston, Massachusetts. They in turn have subcontracted to the Pennsylvania State University Meteorology Department for support work. Pennsylvania State has been a pioneer in the development of the isentrophic trajectory technique, and the personnel working on this Project at both Main and Pennsylvania State have had substantial experience in this area (Figure 6-1).

The project has been designed in two phases with two complimentary approaches. One approach will be to look in detail at 30 of the rain events. A complete work up including precipitation chemistry, trajectory, and meteorological analysis will be performed (called case studies) for each event. Each event for analysis will be selected on the basis of already developed criteria. For example, the lowest pH event at each site, a "normal" meteorological event, or an event with "unusual" chemistry. An extremely high pH event may also prove interesting.

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[a] No parcel heat exchange with the environment.

[b] Process in which entropy is conserved.

The principal trajectory method used will be the isentrophic analysis that inherently incorporates vertical motion. However, in cases of extreme vertical motion such as thunderstorms, (where the adiabatic assumption doesn't hold) the ARL model will be applied to the lower level mixed layer. The approach then applies the individual model that is best suited by the actual meteorological situation.

The selection of events will proceed in two phases. The first phase consists of 5 case study evaluations following two complete quarters of deposition data. The case studies will be completed and a Phase 1 report issued. This report will then be reviewed by the "Independent Assessor" who is under contract to the Department of Natural Resources (Chapter 7). If the Phase 1 report is judged acceptable in terms of its meteorological analysis, then Phase 2 will be authorized. Phase 2 is 25 additional case studies through the remaining year and a half of data. If the trajectory Phase 1 report is judged unacceptable, Phase 2 will be cancelled.

The second approach will be a climatological study. Because only 30 of the expected 150 possible rain events will be analyzed in detail, the more general climatological analysis will be applied to all of the rain event data. Some of the analysis will include variations in the major ions by: Season; Storm Type; Precipitation Type, Rate, and Volume; Characteristic Wind Direction; and Temperature.

Quarterly reports will include the quality assured deposition data, the corresponding meteorological data and analysis for each event, and continuation of the climatological tables. The final report is expected in September, 1984.

Preliminary Findings

Because the Atmospheric Trajectory Study must wait for the deposition data to be supplied, it is necessarily at the end of the line. Only the first two quarterly reports that discuss the first six months of the deposition data are available. Of course, the first quarter of the deposition data includes only Legend Lake and Lake Geneva sites. The Round Lake data will be added to future quarterly reports as the data become available.

The first quarterly report included only a brief synopsis of the chemical data with some simple correlation information. Also, included in this report was a description of the trajectory isentrophic method that is being employed in this project and the first case study that has been accomplished by our consultants. The description of the method utilizes the case study as an example to explain the method. With very little data available for the first quarterly report, little more can be said about the preliminary findings of this study.

The second quarterly report covering the deposition data from June through August of 1982 was not available at the time this report was written, but is expected soon. With this second quarterly report, we will then have enough information to make a selection of the five Phase 1 case studies. Following the selection of these case study events, the Phase 1 report is expected in June, 1983.

7. Trajectory Quality Assurance Project

Introduction

Since atmospheric analysis techniques are complex and the research methods can be interpreted differently, the Joint Acid Deposition Technical Review Committee elected to get a peer review and an independent assessment of the Atmospheric Trajectory Study (Chapter 6).

The objective of the independent assessment project is to provide an independent assessment peer review of the trajectory/climatology study, which insures a high level of credibility and high quality results. The primary tasks of the independent assessor are to:

1) Critique methods used in the Atmospheric Trajectory Study

   a. for trajectory analysis,

   b. for determination of precursor source areas,

   c. for assessing the climatology of acid precipitation in Wisconsin;

2) Describe and use methods to check the meteorological consistency of the computed trajectories;

3) Determine if the proposed methods were properly applied;

4) Evaluate trajectory case studies;

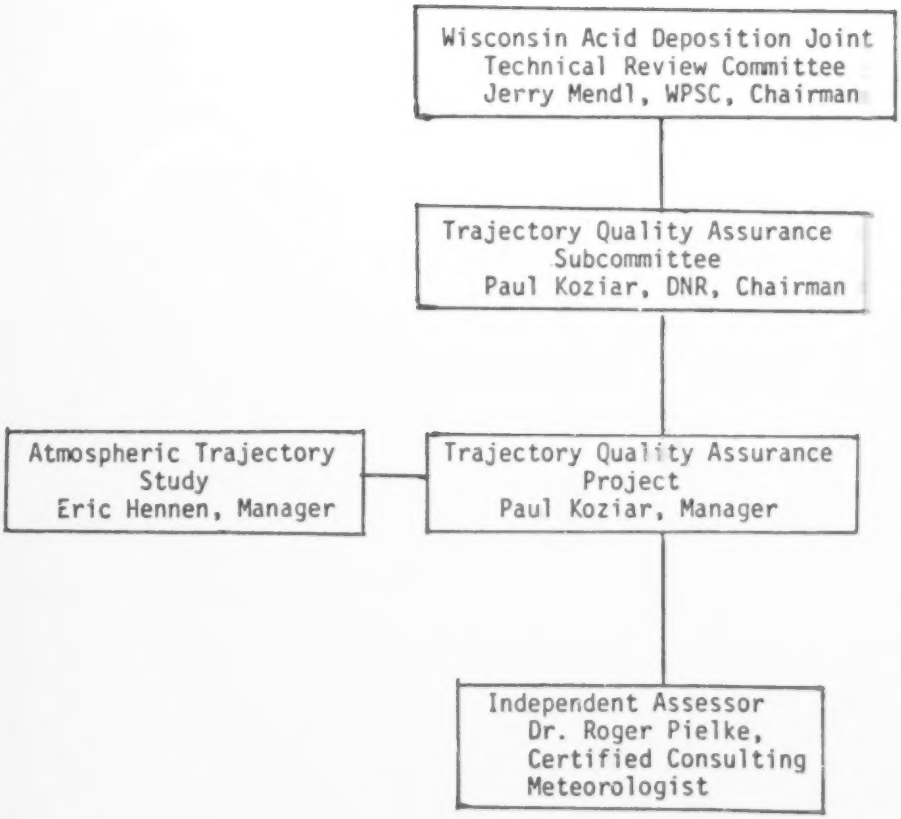
5) Assess the conclusions with regards to trajectories of air parcels and emission source areas responsible for contributing acid precipitation in Wisconsin, the climatology of acid precipitation, and the relationships of the chemical constituents in precipitation with meteorological and emission source factors.

Project Manager for this independent assessment is Paul Koziar, Bureau of Air Management, Wisconsin Department of Natural Resources. Technical Review is provided by the Trajectory Quality Assurance Subcommittee of the Joint Acid Deposition Technical Review Committee with project responsibilities shown in Figure 7-1.

Project History

The project began in October, 1981 when it was determined that an individual with recognized expertise in trajectory studies, dynamic meteorology, and meteorological statistics was needed to conduct the independent assessment of the trajectory work. A literature search was then conducted to compile a list of candidates for the independent assessment contract.

Figure 7-1. Organization and Reporting Structure of the Trajectory Quality Assurance Project


Wisconsin Acid Deposition Joint Technical Review Committee
Jerry Mendl, WPSC, Chairman
Trajectory Quality Assurance Subcommittee
Paul Koziar, DNR, Chairman
Atmospheric Trajectory Study
Eric Hennen, Manager
Trajectory Quality Assurance Project
Paul Koziar, Manager
Independent Assessor
Dr. Roger Pielke, Certified Consulting Meteorologist

In March, 1982, scope of work was drafted and the field of possible contractors was narrowed. In April, 1982, telephone inquiries were made to twelve individuals at research and academic institutions soliciting interest in the project. A scope of work was finalized in May, 1982 and was mailed to seven individuals who had previously expressed interest in the project. Responses to the scope of work were evaluated, and Dr. Roger Pielke of Colorado State University was chosen to conduct the independent assessment on the basis of his expertise, qualifications, the technical merit of his proposal, and the cost of his proposal.

The project will terminate in late 1984 when it is anticipated that Dr. Pielke will make his final report assessing the Phase III Report of the Atmospheric Trajectory Study.

Preliminary Findings

No results are available at this time.

8. Emission Sources Inventory

Introduction

An inventory of sources of acid deposition precursors (primarily sulfur dioxide and nitrogen oxides) is being developed to identify and characterize sources that may contribute to acid deposition in Wisconsin and quantify their emissions. In January 1982 the DNR started to develop a comprehensive inventory including point, line, and area sources for Wisconsin, surrounding states, and southeastern Canadian Provinces. The inventory was designed to include both anthropogenic and natural sources of SO_2 and NO_x (base year 1980 and projections for 1990). The inventory should be completed by July, 1983.

Project Manager for this inventory is Paul Koziar, Bureau of Air Management, Wisconsin Department of Natural Resources. Technical review is provided by the Emission Sources Subcommittee of the Joint Acid Deposition Technical Review Committee with project responsibilities shown in Figure 8-1.

Project Description

The project began in January of 1982 and has been staffed by the Bureau of Air Management. Although the termination date was originally anticipated to be July 1984 it is anticipated that this project will be finished earlier.

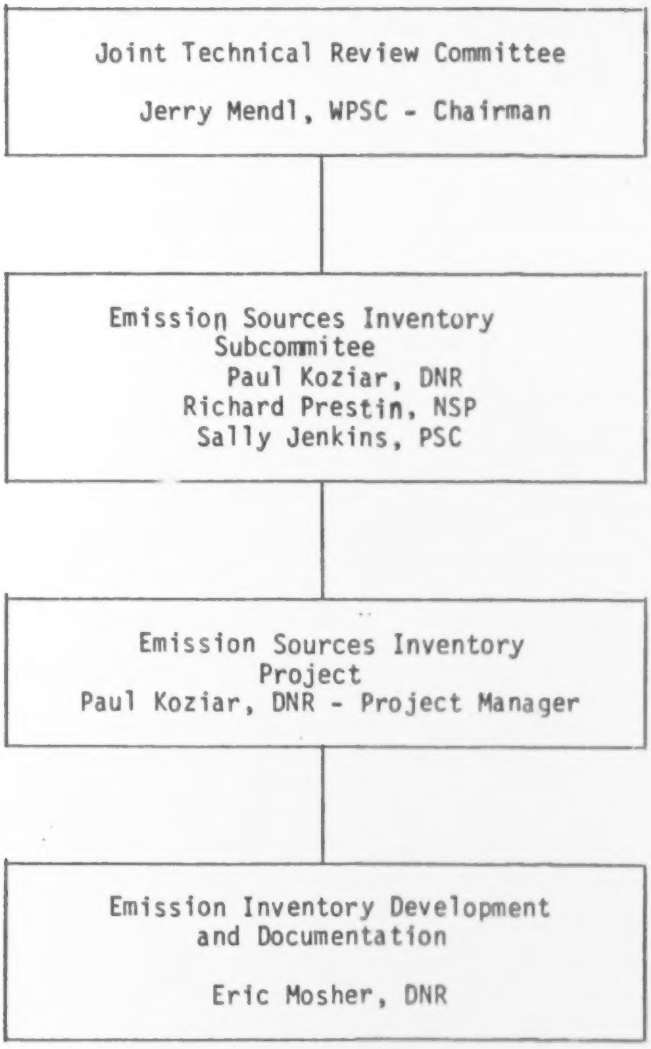
Following is a discussion of work completed to date.

SURE Inventory - The Electric Power Research Institute has previously developed emission inventories of SO_2 and NO_x for the northeastern United States and southeastern Canada for the Sulfate Regional Experiment (SURE). The emissions on the SURE inventory are represented spatially on a rectangular grid with 80 x 80 km grid squares (Figure 8-2). The DNR has chosen to simply extend this grid for use in Wisconsin acid deposition studies (see Figure 13). The services of the U.W. Cartographic Lab have been retained to graphically construct the Wisconsin version of the SURE grid on a universal transverse mercator map projection. The emissions calculated for the SURE inventory were assumed to be correct after a cursory check of the data.

A detailed examination of sources of SO_2 and NO_x inside Wisconsin was made using 1980 as the base year.

Point Sources - Point sources were considered to be stationary sources whose emissions (SO_2 or NO_x) were greater than 10 tons/year. The Wisconsin DNR Air Emission Inventory (NR 101 file) was used as a basis for this inventory.

Figure 8-1. Organization and Reporting Structure of the Acid Deposition Emission Sources Inventory


Joint Technical Review Committee
Jerry Mendl, WPSC - Chairman
Emission Sources Inventory Subcommitee
Paul Koziar, DNR
Richard Prestin, NSP
Sally Jenkins, PSC
Emission Sources Inventory Project
Paul Koziar, DNR - Project Manager
Emission Inventory Development and Documentation
Eric Mosher, DNR

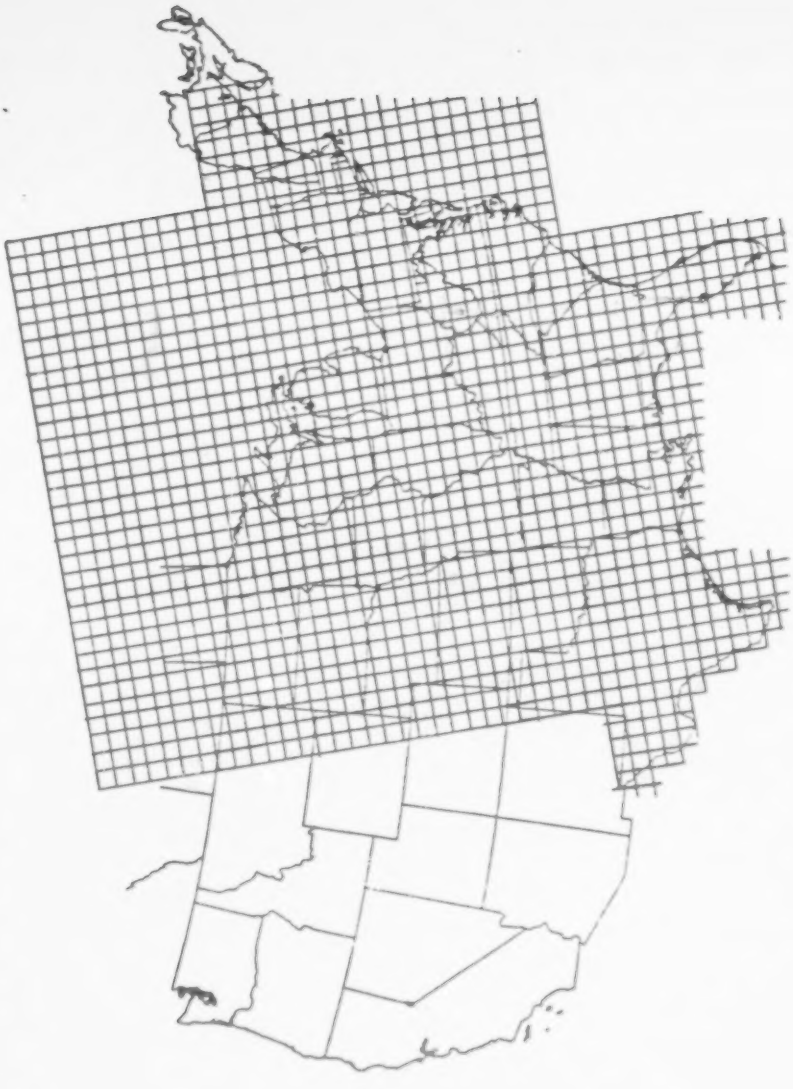


Figure 8-2. Wisconsin Acid Deposition Emission Inventory 80 x 80 Kilometer Grid Squares

Line Sources - Emission factors were obtained from Compilation of Air Pollutant Emission Factors and were applied to state traffic data, which was supplied by the Wisconsin Department of Transportation, to determine SO_2 and NO_x emissions for Wisconsin. The emissions were allocated to counties in the same proportion as the National Emissions Data System (NEDS) 1980 line source inventory for Wisconsin. Emissions for the seven county southeastern corner of the state were computed by the Southeastern Wisconsin Regional Planning Commission.

Area Sources of Anthropogenic Emissions - Emissions from small industrial, commercial, residential, and other small miscellaneous sources of SO_2 and NO_x are considered to be area sources. Anthropogenic area source emissions for most counties were obtained from the National Emissions Data System Area Source Emissions Report for 1980. Emissions from the seven county southeastern corner of the state were computed by the Southeastern Wisconsin Regional Planning Commission.

Area Sources of Natural SO_2 - A literature search was conducted to determine if it was possible to inventory SO_2 from biological processes. It was found that the Electric Power Research Institute, as part of the SURE study, has estimated biogenic SO_2 emissions for all states east of the Mississippi River, including Wisconsin. The SURE emissions estimated for Wisconsin were allocated to counties by relative area.

Area Sources of Natural NO_x - Natural emissions of NO_x include decay of biological material, fertilizer application, animal urine and some agricultural practices. A literature search was conducted to determine if NO_x emissions from natural sources could be inventoried. From the outset of the literature search it was obvious that there would be difficulty in completing the inventory. Emission factors varied an order of magnitude for the same process. Emissions of NH_3 and N_2O complicate the inventory and are often not separated from NO_x emissions. More effort will be made to inventory natural sources of NO_x.

An examination of sources outside of Wisconsin was also made using 1980 as the base year.

U.S. Point Sources - The SURE inventory was relied upon to supply information on sources east of the Mississippi River. Point source data were obtained individually from the states of Texas, Arkansas, Oklahoma, Kansas, Nebraska, Missouri, Iowa, North Dakota, SOuth Dakota, and Minnesota. Emissions were allocated to appropriate grids.

Canadian Point Sources - Information on Canadian sources was obtained from Environment Canada and the provinces of Manitoba, Quebec and Ontario. Emissions will be allocated to appropriate grids.

U.S. Line and Area Sources - The SURE inventory was relied upon to supply data on area and line source emissions for states east of the Mississippi River. For states west of the Mississippi, area and line source data were obtained from the National Emissions Data System Inventory.

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Canadian Line and Area Sources - A gridded emissions inventory from Environment Canada was used to compile Canadian line and area source emissions.

Natural Sources - The Environment Canada and NEDS inventories do not include natural sources that must be inventoried separately.

SO_2 - U.S. natural sources of SO_2 were inventoried similarly to Wisconsin natural sources. Canadian sources are yet to be inventoried.

NO_x - Natural sources of NO_x have not been inventoried because of the previously mentioned problems.

Preliminary Findings

For sources inside Wisconsin, the following conclusions have been made.

1) Point sources of sulfur dioxide have been categorized. Tables 8-1, 8-2, and 8-3 and Figures 8-3 and 8-4 illustrate some of the results. Generation of electric power is the most significant source of SO_2 in the state. Pulp and paper mills are the next most important source. Only 13 sources represent 80% of the total SO_2 emissions in Wisconsin in 1980.

2) Line sources represent only about 1% of the total emissions of SO_2 in the state.

3) Area sources, both natural and anthropogenic, represent a very small fraction (0.7%) of the total SO_2 emissions in Wisconsin. Some particular sources such as bacterial decomposition of asphalt and sulfur dioxide emitted from paving were investigated in some detail and were determined to be negligible as sources of SO_2.

4) Point sources for nitrogen oxides have been categorized. Tables 8-1, 8-4, and 8-5 and Figures 8-5 and 8-6 illustrate some of the results. Generation of electric power was the most significant source category of NO_x emissions. Pulp and paper production was again second. Only 24 sources of NO_x represent 88% of the point source emissions in the state.

5) Line sources proved to be a significant contributor to the total NO emissions in the state (40-50% of anthropogenic emissions).

6) Area sources of NO_x are only a minor contributor to the total anthropogenic emission in the state.

7) Natural sources of NO_x proved to be extremely difficult to inventory and will continue to be investigated as best possible in order to estimate NO_x emissions in the state.

For sources outside of Wisconsin a detailed categorization of emission sources has not been completed. However, certain generalities, such as the insignificance of natural sources of SO_2, may be extrapolated to other areas in the United States and Canada. Line sources of NO_x are certainly significant especially in highly populated areas.

Table 8-1. Wisconsin 1980 Point Source SO_x and NO_x Emissions by Source Size Category

SO_x

Source Size Category (tons/yr)	Number of Sources	%	SO_x Emissions (tons/yr)	%
> 10,000	13	2.4	549,594	79.6
1000-10,000	30	5.6	121,883	17.6
100-1000	40	7.5	16,309	2.4
10-100	76	14.3	2,390	0.3
0.1-10	374	70.2	727	0.1
Total	533	100.0	690,903	100.0

NO_x

Source Size Category (tons/yr)	Number of Sources	%	NO_x Emissions (tons/yr)	%
> 10,000	5	0.6	103,685	51.8
1000-10,000	19	2.3	73,150	36.5
100-1000	62	7.6	15,832	7.9
10-100	201	24.6	6,136	3.1
0.1-10	531	64.9	1,528	0.7
Total	818	100.0	200,331	100.0

Table 8-2. Wisconsin Point Sources Which Emitted Over 1000 Tons Of Sulfur Oxides In 1980

Rank	Facility ID NO.	Facility Name	Facility Location	1980 SO_x Emissions (Tons)[a]				
				1	2	3	4	Annual
1	410054	WEPCo Oak Creek	Oak Creek	26,255	26,255	22,675	44,156	119,340
2	600028	WPSL Edgewater	Sheboygan	18,127	11,695	15,788	12,864	58,474
3	630003	Dairyland Power	Genoa	16,895	14,639	14,975	10,595	56,301
4	50013	WPSC Pulliam	Green Bay	13,743	11,915	10,541	9,624	45,824
5	111007	WP&L Columbia	Portage	9,211	10,986	11,305	10,467	41,969
6	450016	WEPCo Port Washington	Port Washington	12,361	10,303	9,479	9,066	41,211
7	410032	WEPCo Valley Plant	Milwaukee	11,900	9,359	11,394	9,138	40,691
8	60001	Dairyland Power	Alma	6,493	5,483	10,578	10,578	34,122
9	220001	WP&L Nelson Dewey	Cassville	10,072	7,869	5,665	7,869	31,475
10	50003	Proctor & Gamble Fox River Mill	Green Bay	6,008	6,008	6,008	6,008	24,032
11	370003	WPSC Weston Plant	Weston	6,554	6,101	4,972	4,972	22,599
12	50008	Fort Howard Paper Co.	Green Bay	4,477	4,477	4,477	4,477	17,908
13	540003	WPSL Rock River	Beloit/Janesville	6,140	2,834	2,676	4,093	15,743
14	720011	Nekoosa Papers, Inc.	Port Edwards	2,383	2,383	2,383	2,383	9,532
15	450011	Thilmany Pulp & Paper	Kaukauna	2,183	2,184	2,183	2,183	8,733
16	720012	Nekoosa Papers, Inc.	Nekoosa	2,178	2,179	2,179	2,179	8,715
17	720009	Consolidated Papers, Inc.	Biron	2,143	2,143	2,143	2,143	8,572
18	130011	Madison Gas & Electric	Madison	2,946	1,800	2,045	1,391	8,192
19	440001	Rhinelander Paper Co.	Rhinelander	1,663	1,664	1,563	1,664	6,654
20	720010	Consolidated Papers Kraft Div.	Wisc. Rapids	1,662	1,661	1,662	1,561	6,646
21	350004	Owens-Illinois Forest Prod. Division	Tomahawk	1,385	1,500	1,384	1,500	5,769
22	450008	Midtec Paper Corp.	Kimberly	1,073	1,073	1,340	1,877	5,363
23	300003	WEPCo Pleasant Prairie	Kenosha	0	0	2,229	3,078	5,307
24	220002	Dairyland Power Stoneman Sta.	Cassville	2,700	778	208	1,506	5,192
25	370055	Weyerhaeuser Company	Rothschild	1,283	1,170	1,123	1,123	4,699
26	370004	Wausau Paper Mills	Brokaw	905	904	905	904	3,619
27	380004	Niagara of Wisc. Paper Co.	Niagara	868	868	868	868	3,472
28	20004	Bay Front Generating Sta	Ashland	806	687	717	778	2,988
29	130029	UW-Madison	Madison	800	585	559	746	2,690
30	450004	Consolidated Papers, Inc.	Appleton	663	663	662	663	2,651
31	370009	Mosinee Paper Company	Mosinee	661	661	661	661	2,644
32	710006	Menasha Corp. Paperboard Mill	Menasha	591	592	591	592	2,366
33	540012	WP&L Blackhawk Station	Beloit	860	156	645	294	1,955
34	380002	Badger Paper Mills, Inc.	Peshtigo	468	468	468	468	1,872
35	410054	Milwaukee Co. Institutions	Wauwatosa	492	401	455	474	1,822
36	510001	Flambeau Paper Corp.	Park Falls	479	479	373	444	1,775
37	540004	General Motors Assembly Division	Janesville	402	490	385	472	1,749
38	130012	Oscar Mayer & Co.	Madison	402	402	453	420	1,677
39	160012	Murphy Oil Corp.	Superior	412	411	412	411	1,646
40	720001	Marshfield Electric	Marshfield	434	338	402	434	1,608
41	50010	Green Bay Packaging	Green Bay	349	349	349	349	1,396
42	50011	Nicolet Paper Co.	DePere	326	327	326	327	1,306
43	360006	Manitowoc Public Utilities	Manitowoc	325	326	325	326	1,302
Total				179,950	156,465	159,730	175,329	671,474

a. Quarterly and annual emissions

Table 8-3.

Wisconsin 1980 Point Source SO_x Emissions By Source Category and Fuel Type
(includes only the 43 sources which emitted over 1000 tons)

Emission Source	1980 SO_x Emissions(tons)				Percent of Total 1980 Point Source SO_x Emissions (690,903 tons)
	Electric Utility Generating Stations	Paper Mills	Others	Total	
Coal	533,976	80,501	7937	622,414	90.1
Pulping Liquor	0	19,484	0	19,484	2.8
Residual Oil	33	5566	326	5925	0.9
Wood	57	427	0	484	-
Distillate Oil	121	6	1	128	-
Refinery Gas	0	0	61	61	-
Total Boiler Emissions	534,187	105,984	8325	648,496	93.9
Process Emissions	0	21,719	1259	22,978	3.3
Total Emissions	534,187	127,703	9584	671,474	97.2
Percent of Total 1980 Point Source SO_x Emissions	77.3	18.5	1.4	97.2	-

Figure 8-3. Location of Point Sources in Wisconsin Which Emittad Over 1,000 Tons of Sulfur Dioxide in 1980

Figure 8-4.
Wisconsin 1980 Sulfur Dioxide Emissions by County

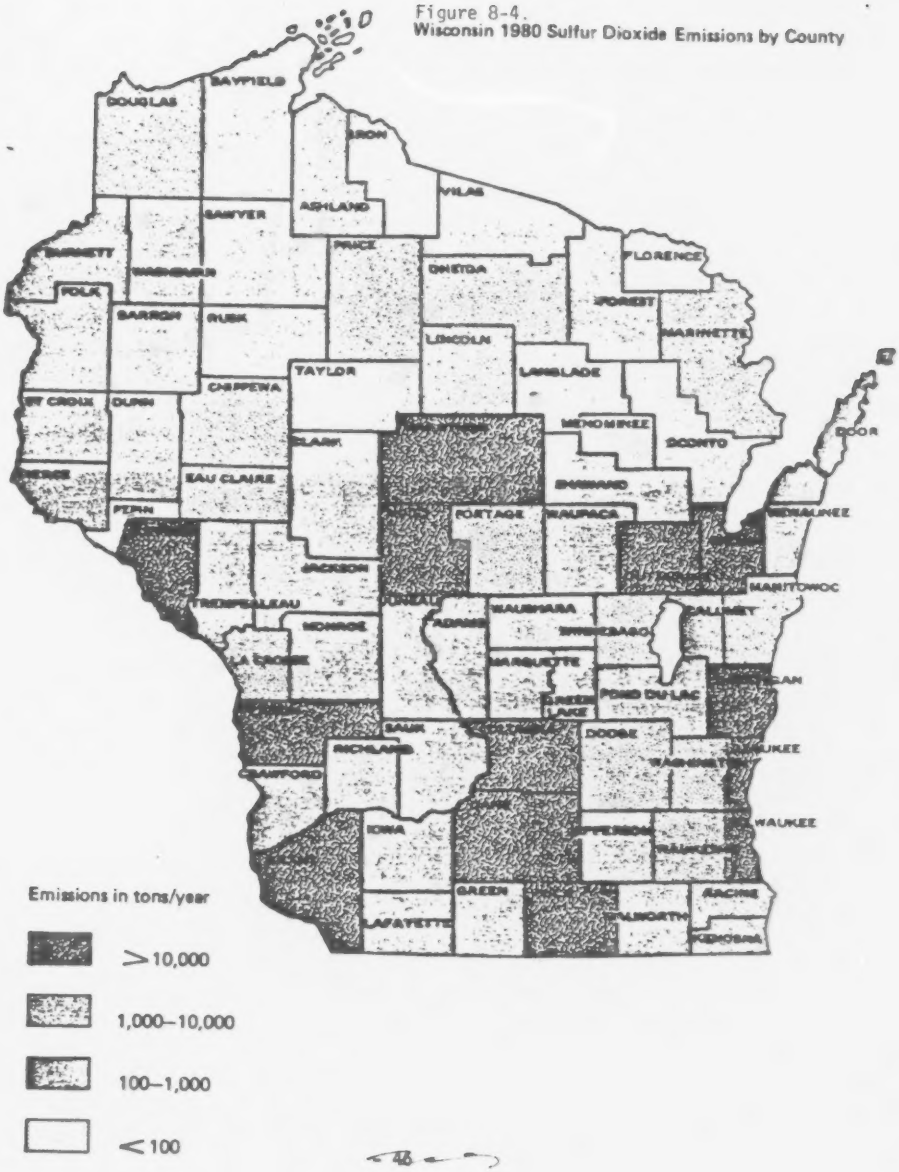


Table 8-4.

Wisconsin Point Sources Which Emitted Over 1000 Tons of Nitrogen Oxides in 1980

Rank	Facility ID No.	Facility Name	Facility Location	1980 NO_x Emissions (Tons)				
				Winter	Spring	Summer	Fall	Annual
1	600008	WP&L Edgewater	Sheboygan	8906	5746	7757	6321	28,730
2	110007	WP&L Columbia	Portage	5717	6757	7016	6497	25,987
3	410094	WEPCo Oak Creek	Oak Creek	5066	5066	4376	8520	23,028
4.	220001	WP&L Nelson Dewey	Cassville	4860	3796	2733	3797	15,186
5.	60001	Dairyland Power	Alma	2043	2043	3334	3334	10,754
6.	630003	Dairyland Power	Genoa	2472	2142	2060	1566	8,240
7.	50013	WPSC Pulliam	Green Bay	2154	1867	1652	1508	7,181
8.	410103	WEPCo Valley Plant	Milwaukee	2030	1611	1961	1401	7,003
9.	540003	WP&L Rock River	Beloit Janesville	2677	1236	1167	1784	6,864
10.	50008	Fort Howard Paper Co.	Green Bay	1549	1549	1549	1549	6,196
11.	460016	WEPCo Port Washington	Port Washington	1846	1538	1415	1353	6,152
12.	300009	WEPCo Pleasant Prairie	Kenosha	0	0	2199	3036	5,235
13.	450011	Thilmany Pulp & Paper	Kaukauna	992	991	992	991	3,966
14.	350004	Owens-Illinois Forest Prod.	Tomahawk	738	800	739	800	3,077
15.	370003	WPSC Weston Plant	Weston	860	801	652	652	2,965
16.	440001	Rhinelander Paper Co.	Rhinelander	712	713	713	713	2,851
17.	720009	Consolidated Papers, Inc.	Biron	662	662	662	663	2,649

Table 8-4 (cont.)
Wisconsin Point Sources Which Emitted Over 1000 Tons of Nitrogen Oxides in 1980
(Page 2)

Rank	Facility ID No.	Facility Name	Facility Location	1980 NO_x Emissions (Tons)				
				Winter	Spring	Summer	Fall	Annual
18.	20004	Lake Superior Dist Power Co. Bay Front Generating Sta.	Ashland	591	504	526	569	2,190
19.	50003	Procter & Gamble Fox River Mill	Green Bay	510	511	510	511	2,042
20.	720010	Consolidated Papers, Inc. Kraft Division	Wisconsin Rapids	410	409	410	409	1,638
21	130011	Madison Gas & Electric	Madison	523	319	363	247	1,452
22.	720012	Nekoosa Papers, Inc.	Nekoosa	309	310	310	310	1,239
23.	410095	WEPCo Lakeside Sta.	Milwaukee	484	231	242	196	1,153
24.	450008	Midtec Paper Corp	Kimberly	211	212	264	370	1,057
Total				46,322	39,814	43,602	47,097	178,835

Table 8-5. Wisconsin 1980 Point Source NO_x Emissions
(includes only the 24 sources which emitted over 1000 tons)

Emission Source	1980 NO_x Emissions (tons)			Percent of Total 1980 Point Source NO_x Emissions (200,331 tons)
	Electric Utility Generating Stations	Paper Mills	Total	
Coal	149,930	21,664	171,594	85.7
Wood	382	1869	2251	1.1
Natural Gas	1560	128	1688	0.8
Pulping Liquor	0	802	802	0.4
Distillate Oil	224	3	227	0.1
Residual Oil	24	105	129	0.1
Total Boiler Emissions	152,120	24,571	176,691	88.2
Process Emissions	0	144	144	0.1
Total Emissions	152,120	24,715	176,835	88.3
Percent of Total 1980 Point Source NO_x Emissions (200,331 tons)	75.9	12.4	88.3	—

Figure 8-5. Location of Point Sources in Wisconsin Which Emitted Over 1,000 Tons of Nitrogen Oxides in 1980

Figure 8-6.
Wisconsin 1980 Nitrogen Oxides Emissions by County

9. Intensive Watershed Study

Introduction

This project is a study of two lake watersheds in northwest Wisconsin, organized as a joint effort among researchers at the University of Wisconsin, the U.S. Geological Survey, and the Wisconsin Department of Natural Resources. Prior to final selection of study lakes, an extensive screening process was employed based on a number of established criteria believed to be important to meet study objectives. Because of other ongoing studies in north central Wisconsin, the Northwest region was chosen for this study. Desirable lake characteristics sought included apparent high and medium susceptibility (low and medium alkalinity) to acid deposition, small size (ca. 25-100 acres), no stream inlets or outlets, fish populations typical of the region, good access, and geologic suitability for development of a hydrologic budget. Eliminated as undesirable for study sites were darkstained bog lakes, lakes which might go dry, lakes subjected to any form of chemical management, and lakes that have winter fish kills.

The two lake watersheds selected for study were Round Lake, Douglas County and East Eight Mile Lake, Bayfield County (Figure 9-1). The study sites both lie in the Northern Highland physiographic province of Wisconsin; general susceptibility of soils and lake waters in the region to changes in pH by acid deposition are shown in Figures 9-2 and 9-3. One susceptible lake (Round) and one potentially susceptible lake (East Eight Mile) were chosen to allow comparison of the structure and function of two ecosystems with differing responses to similar atmospheric inputs. The lakes are approximately 25 miles apart. The watersheds include soils and vegetation types commonly found in northwestern Wisconsin, and hydrologically are typical of other landlocked lakes in the region.

Specific objectives of this watershed research project are as follows:

1) To determine the quantity and quality of atmospheric deposition in the watersheds.

2) To assess the modification of precipitation by various forest cover types, and describe the modification of the water as it passes through the soils.

3) To determine the hydrologic budgets and assess the impact of budget components on lake-water quality.

4) To investigate the impact of acid deposition on lake biota.

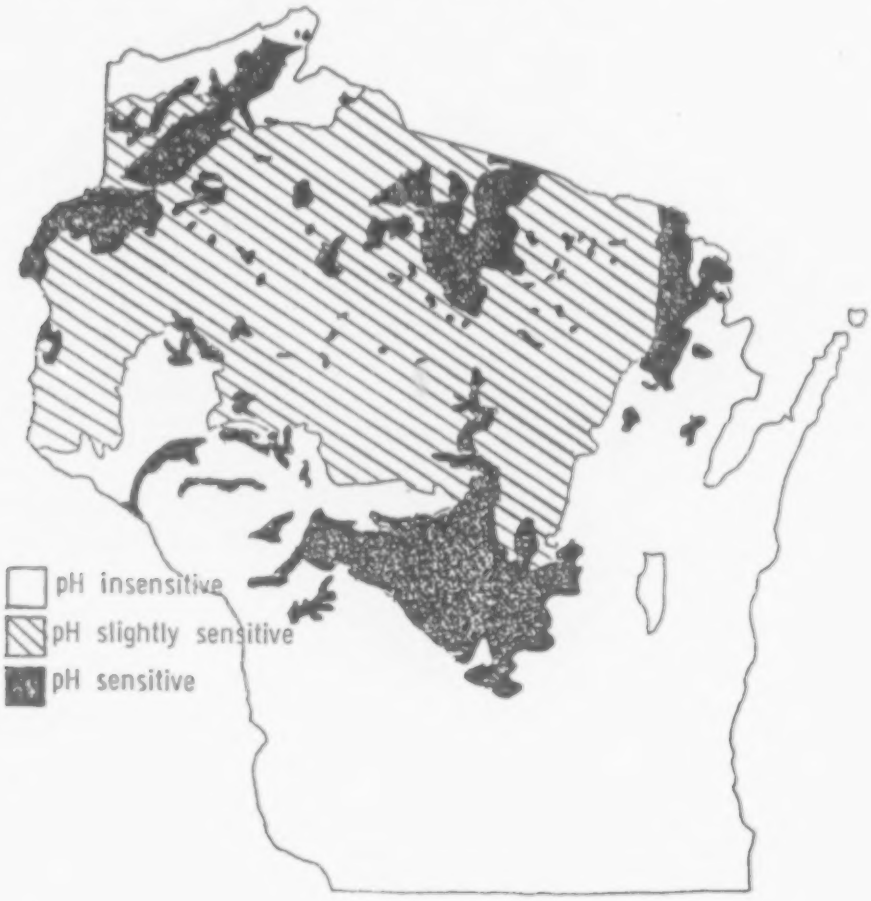
5) To evaluate the pathological effects of acid precipitation on the foliage of vegetation.

6) To integrate the research findings by two methods, i.e. elemental flux calculations and mathematical modeling.

7) To adapt, calibrate, and verify the ILWAS model to the system to facilitate prediction.

Figure 9-1. Location of Study Areas in Wisconsin

Figure 9-2. Susceptibility of surface mineral soil by soil region in Wisconsin to change in pH by acid precipitation



J. G. Bockheim, UW-Madison, 1980

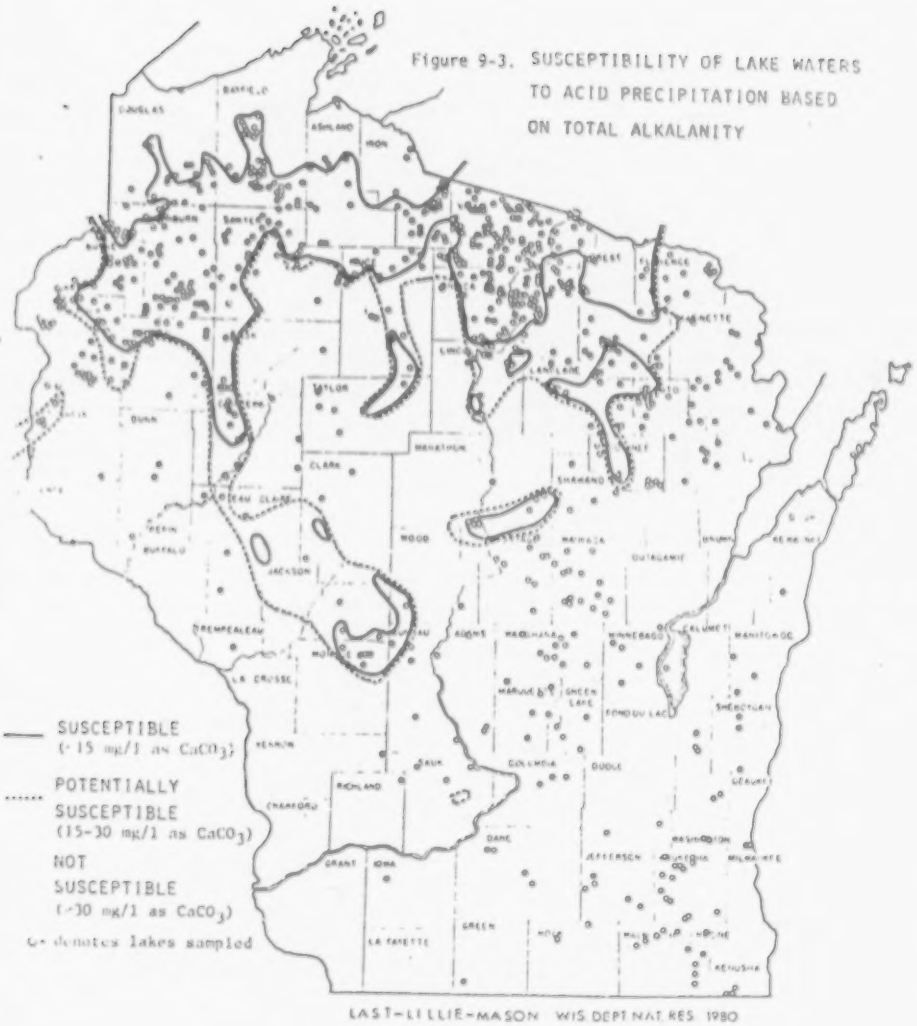


Figure 9-3. SUSCEPTIBILITY OF LAKE WATERS TO ACID PRECIPITATION BASED ON TOTAL ALKALANITY

Field data collections were fully underway on both study watersheds by August, 1981. The project consists of work in four segments; atmospheric input, terrestrial system, basin hydrology, and limnology.

Atmospheric input is being measured by the Deposition Monitoring Project described in Chapter 4. In addition, deposition samples are being collected as part of the terrestrial system and basin hydrology components. The terrestrial system is being studied by James Bockheim and researchers from the University of Wisconsin-Madison Soils and Forestry Department. Forest pathology is being studied by Allen Prey from the Wisconsin Department of Natural Resources, Bureau of Forestry. The basin hydrology component is being conducted by Dennis Wentz and colleagues from the United States Geological Survey, Water Resources Division. Paul Garrison and staff of the Wisconsin Department of Natural Resources Bureau of Research are conducting the limnological study of the lake. John Mason, Wisconsin DNR, has acted as project coordinator since Thomas Wirth retired from state service in October, 1982. Doug Knauer has since replaced Tom Wirth and now serves as project coordinator.

Modeling of the two lake systems is being carried out by Tetra Tech, Inc. under the direction of Dr. Carl Chen. The ILWAS model, developed for the Integrated Lake Watershed Acidification Study in New York State sponsored by the Electric Power Research Institute (EPRI), is in the process of modification, calibration and verification using Wisconsin watershed data. Technical review for the study is provided by the Watershed Study Subcommittee of the Joint Acid Deposition Technical Review Committee, with project responsibilities shown in Figure 9-4.

Terrestrial Component

Following is a summary of the progress and information gathered in the watersheds of the two study sites.

Acid deposition is occurring at Round and East Eight Mile Lakes, northwestern Wisconsin. During the period August, 1981 until the present, pH of bulk precipitation at both lakes averaged 4.6. The dominant cation and anion is hydrogen and sulfate, respectively.

Predominant forest types at Round Lake are northern pin oak (Quercus ellipsoidalis E. J. Hill) - bur oak (Q. macrocarpa Michx.), jack pine (Pinus banksiana Lamb.), red pine (P. resinosa Ait.), and quaking aspen (Populus tremuloides Michx.). At East Eight Mile Lake, predominant forest types are jack pine, mixed oaks, aspen, red pine, and paper birch (Betula papyrifera Marsh.)

Although density and site index of each of the major forest types are greatest at East Eight Mile Lake, average stand age and basal area are greatest at Round Lake. Site index of major species at Round Lake include jack pine 14.7 m and northern pin oak 7.4 m. At East Eight Mile Lake, site index of jack pine is 20.9, northern pin oak is 8.8, and quaking aspen is 24.8 m.

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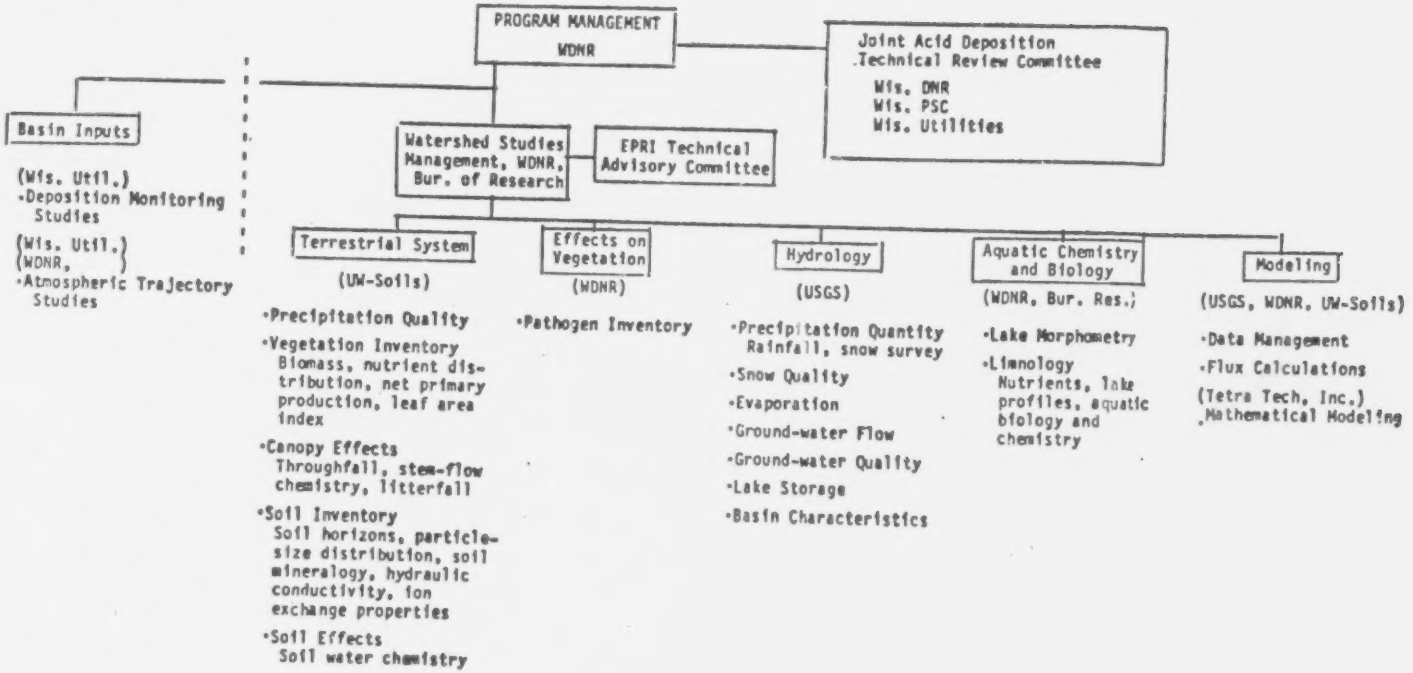
Figure 9-4. Project Management of the Intensive Watershed Study.



Average biomass of jack pine, northern pin oak, red pine and bigtooth aspen are greatest at Round Lake because the stands are older than at East Eight Mile Lake. At Round Lake average biomass for the various forest types was 185 t/ha for red pine, 143 t/ha for bigtooth aspen, 87 t/ha for jack pine, and 52 t/ha for mixed oaks. At East Eight Mile Lake, average biomass was 131 t/ha for paper birch, 91 t/ha for bigtooth aspen, 87 t/ha for quaking aspen, 76 t/ha for jack pine, 40 t/ha for red pine, and 24 t/ha for mixed oaks. Net primary production was low for all species except quaking aspen at East Eight Mile Lake (12.9 t/ha/yr) and red pine at Round Lake (18.2 t/ha/yr). Whereas growth of the aspen stand was influenced by a calcareous subsoil, the red pine stand was influenced by a seasonally high water table. Nutrient contents in the vegetation were dependent on species and stand age. When normalized to an age of 55 years, amounts of N, P, K, Ca, and Mg were greatest in the biomass of quaking aspen, followed by red pine, paper birch, and jack pine.

During 1981 and 1982, 741 vegetation samples were collected from plots located in the Round and East Eight Mile Lake catchment areas that showed signs or symptoms of pathogenic fungi or insect injury. These vegetation samples currently are being examined to determine cause of injury.

The predominant soil on both watersheds is the Omega loamy sand, a sandy, mixed, frigid Typic Udipsamment. This soil occurs uniformly on slopes ranging from 0 to 30%. The profile of the Omega soil has a 1 to 10 cm duff mull forest floor over an organic-enriched A horizon, a weakly expressed E horizon, and a series of color Bw horizons to a depth of 47 to 71 cm. The parent material is acid, sandy outwash of Valderan (ca. 10,000 yr BP) age. The maximum depth of rooting ranges between 54 and 77 cm. The Omega loamy sand is very strongly acid (pH 4.5 to 5.0) in the surface mineral horizon and medium acid (pH 5.6 to 6.0) in the subsoil. Concentrations of organic C, total organic N, acid-extractable P and S, and exchangeable Ca, Mg, and K are very low. All of the extractable elements except P and S decrease with soil depth. Extractable P and S reach maxima in the upper Bw horizon. The Omega soil has a cation-exchange capacity (CEC) of 14 to 21 meq/100 g soil in the surface mineral horizon and a CEC of 0.6 to 4.8 in the subsoil. C:N ratios commonly range between 10 and 20. Saturated hydraulic conductivities are very rapid, commonly ranging between 100 and 600 cm/hr. The proportion of sand in the 2 mm fraction ranges between 85 and 99%. Medium (0.5-0.25 mm) and fine (0.25-0.10 mm) fractions are dominant. Predominant minerals in the clay-size fractions (2-0.2, 0.2-0.08, and $<$0.08 m) are vermiculite, montmorillonite, quartz, and hydrous mica, with smaller amounts of kaolinite, orthoclase feldspar, and plagioclase feldspars. Hydrous mica and chlorite have weathered to form montmorillonite and vermiculite.

Preliminary estimates indicate that the absolute amounts (kg/ha) of Ca, Mg, and K are less in the upper 100 cm of the Omega soil beneath mixed oak than beneath jack pine and that these differences may be attributable to greater uptake of these cations by the oak.

Seasonal fluctuations in soil temperatures at 0, 50, and 100 cm were similar beneath mixed oak and jack pine at Round Lake. Surface soil temperatures were highest in July (22°C). From ca. November 1 until ca. April 1, soil temperatures were inverted, being highest at 100 cm and lowest at 0 cm. The surface soil remained below 0°C between November 1, 1981 until early April 1982 (approximately 133 days). At 50 cm the soil temperature remained below freezing from early January until mid-April (approximately 100 days). The soil did not freeze in 1981-82 at a depth of 100 cm.

Gravimetric soil moisture at Round Lake ranged narrowly between 3.4 and 11.0% (w/w). Soil moisture was slightly lower beneath the jack pine stand than the oak stand, perhaps because of greater canopy interception by the jack pine. The moisture content never reached the permanent wilting point of 15 bars.

Maximum leaf area indices (LAI) occurred approximately two weeks after leafing out on about June 1, 1982. LAI was considerably lower for the deciduous types, including paper birch (1.0 m^2/m^2), quaking aspen (1.1), and mixed oak (1.2) than for jack pine (6.3 to 10.2 m^2/m^2). On October 14, 1982, LAI of deciduous types were 0 and LAI for jack pine ranged between 4.2 and 9.2 m^2/m^2) on the experimental plots.

Despite the lower LAI values, the oak canopy was more efficient in adsorbing H+ ions and returning Ca^{2+} and K^+ in throughfall than the jack pine. Since the jack pine foliage would be expected to trap more dryfall than the oak, these differences are attributed primarily to foliar leaching. Both canopies adsorbed NH_4+, SO_42- and NO_3- from the bulk precipitation.

On both watersheds, jack pine had the greatest litterfall, followed by paper birch, quaking aspen, and mixed oak. After one year, 78 and 77% of the jack pine and mixed oak litter remained following decomposition. After one year of decomposition of mixed oak litter at Round Lake, there was a net gain in N, S, and Ca, and a net loss in K, P, and Mg. The order of mobility of the various elements during decomposition of mixed oak litter was K > P, Mg > Ca, S, N. After one year of decomposition of jack pine litter at Round Lake, there was a net gain in N, S, P, and Ca and a net loss in K and Mg. The order of mobility during decomposition of jack pine litter was K > Mg > Ca, P, S, N.

Preliminary estimates of nutrient uptake suggest that the mixed oak forest type took up nearly twice as much K and 50% greater amounts of Ca and Mg than the jack pine.

Tagged white pine trees in the Round Lake watershed did not show signs and symptoms of air pollution injury during 1981 or 1982.

Whereas loadings of H+, NH_4+, and NO_3- were diminished in the shallow soil percolate beneath mixed oak and jack pine at Round lake, loadings of Ca^{2+}, K+, Mg^{2+}, and SO_42- increased relative to that in throughfall + stemflow. These differences are attributed to fine root turnover and breakdown of soil organic matter.

Leaching losses at a soil depth of 60 cm were greater for N (NH_4^+ + NO_3^-) and Ca^{2+} beneath jack pine than mixed oak at Round Lake. There were no significant differences in leaching losses of K^+, Mg^{2+}, and SO_4^{2-}. Leaching losses were 330 meq/ha/yr and were similar to values reported in the literature for Quercus-Pinus ecosystems.

Although our data and data from the literature show increased loading of H^+, NH_4^+, and SO_4^{2-}, NO_3^-, and perhaps other chemical constituents due to anthropogenic activities, we are unable to detect effects of this increased loading on vegetational and soil properties and processes. However, we can show differences in nutrient fluxes and pools due to differences in forest types in as little as 30 to 50 years.

Hydrologic Component

The following section addresses the progress of the hydrologic investigation from the beginning of data collection in August 1981 through at least September 1982. In many cases, data collected beyond September 1982 are discussed, and often the ending dates for various discussions are not consistent. This is because the data are received and processed on different schedules, and the data-base management programs which summarizes the data, are completed at different times. Thus, the discussions include the most recent data to have been processed at the time that section was written.

Included in the text that follows are (1) a brief discussion of the locations of the sites for collection of hydrologic and water-quality data, and (2) a discussion of preliminary results, including some tentative interpretations of these results.

Hydrologically, the lakes and their surroundings seem to react in much the same manner. Precipitation, evaporation, and changes in groundwater levels and lake stage are comparable at the two study areas. Measurements of specific hydrologic variables generally correlate well, thus allowing missing data at one study area to be estimated from data collected at the other study area.

Water quality of snow collected from both watersheds is similar. Changes during snowmelt indicate the same processes are controlling. Data showing that 51 to 63 percent of the ion load was released with the first 26 to 38 percent of the melt water during March 1982 are consistent with data reported in the literature.

The quality of the deep groundwater is similar at the two study areas. Concentrations of Ca, Mg, and alkalinity are approximately twice those in the shallow groundwater. Except for one deep piezometer at Round Lake, the deep groundwater from both study areas is at or near saturation with respect to calcite.

The most obvious difference between the two study areas is the quality of the shallow groundwater. Concentrations of most constituents are higher at East Eight Mile Lake, particularly in the discharge area along the south shore. This is the only area where the shallow groundwater is saturated with respect to calcite.

Locations of the hydrologic instrumentation and water quality sampling sites are shown on Figure 9-5 for Round Lake and on Figure 9-6 for East Eight Mile Lake. A brief description of the hydrologic instrumentation and water quality sampling equipment, and information regarding the spatial and temporal distribution of data collection are given in Tables 9-1 and 9-2.

The hydrologic budget of a typical lake is shown diagrammatically on Figure 9-7. The equation describing the interrelationships of such a budget is:

$$\Delta S = P - E \pm G + SF + OF - O, \quad (1)$$

where

ΔS is change in lake storage,

P is precipitation on the lake surface,

E is lake evaporation,

G is groundwater recharge from or discharge to the lake,

SF is streamflow to the lake,

OF is overland flow to the lake by ephemeral channelized or by nonchannelized surface runoff, and

O is water lost from the lake via a surface outlet.

Round and East Eight Mile Lakes are both seepage lakes, and the streamflow and lake-outlet components do not exist. In addition, because the lakes are situated in sandy outwash and are surrounded by mature forests, overland flow is thought to be negligible. Thus, equation 1 reduces to

$$\Delta S = P - E \pm G. \quad (2)$$

The approach to determining the hydrologic budget at Round and East Eight Mile Lakes is to measure each of the components in equation 2. Because each term in the equation has associated measurement and interpretation errors (Winter, 1981), a better representation of the hydrologic budget is given by:

$$\Delta S = P - E \pm G \pm e, \quad (3)$$

where e is the overall error term. By measuring S, P, E, and G in equation 3, an estimate of e can be calculated. This provides one criterion for determining the worth of the hydrologic budget.

Estimates of the various terms in equation 3 for Round and East Eight Mile Lakes have not yet been determined; however, some general comments regarding the various budget components follow.

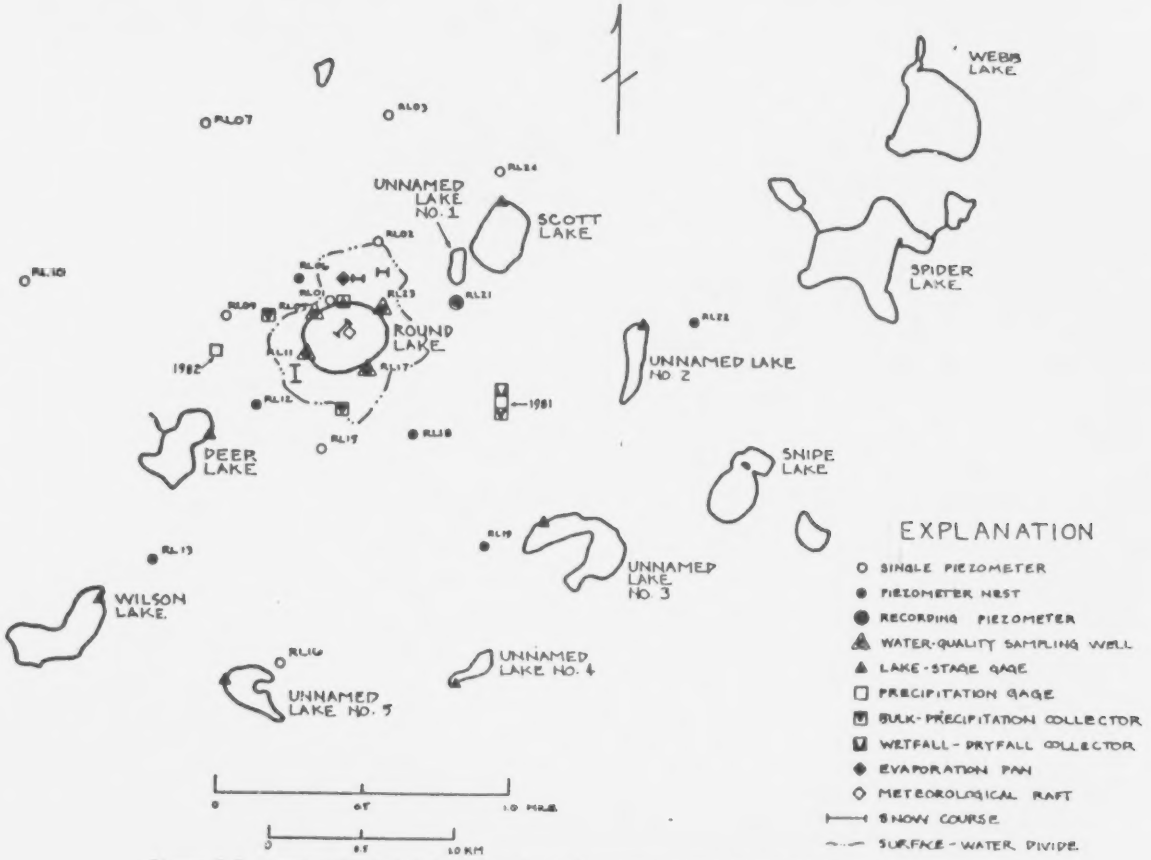


Figure 9-5. Location of hydrologic instrumentation and water-quality sampling sites at the Round Lake study area.

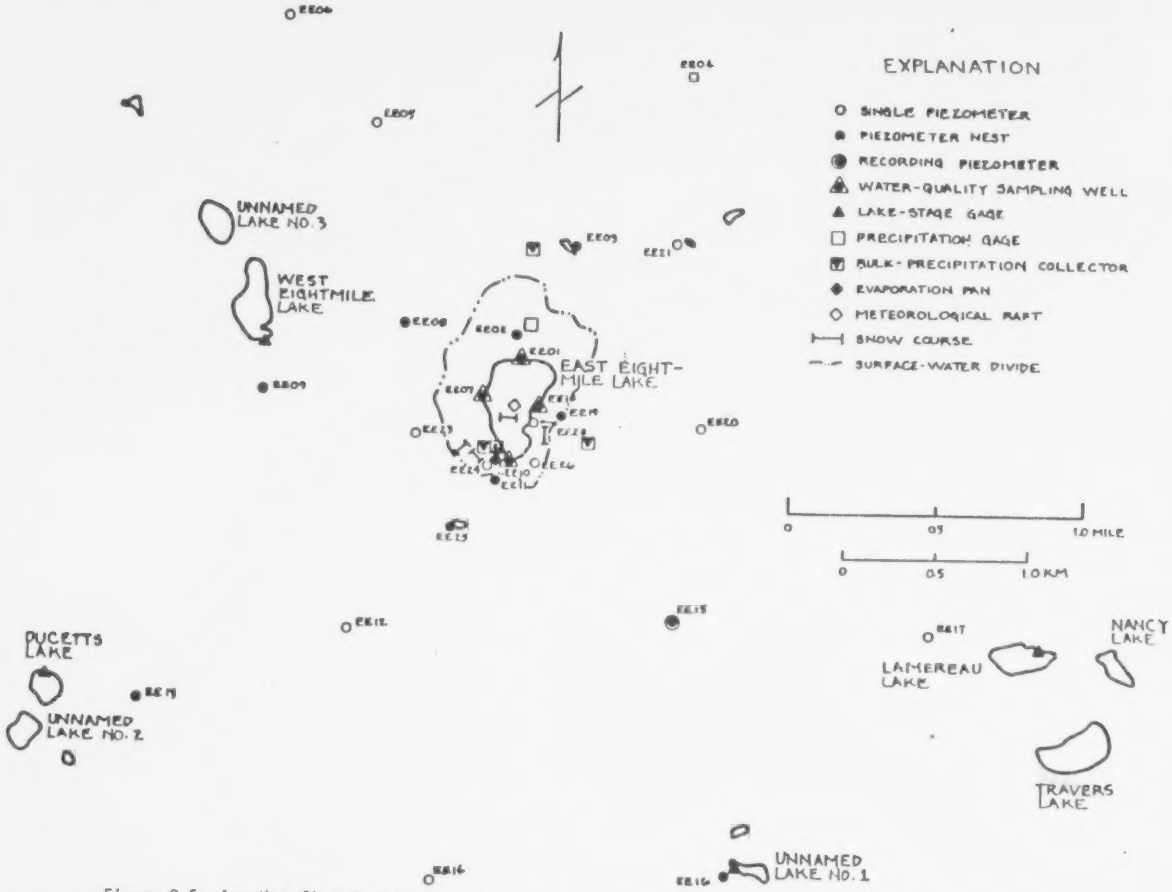


Figure 9-6. Location of hydrologic instrumentation and water-quality sampling [illegible]

Table 9-1. Description of instrumentation for hydrologic-budget data collection

Hydrologic-budget component	Instrumentation	Number per watershed	Frequency of collection	Period of collection
Lake storage	Lake-stage gage	1	Continuous	Year-round
Precipitation	200-mm (8-in.) gage	1	Continuous	Nonfreezing periods
	200-mm (8-in.) gage	1	Daily	Year-round
	Snow courses	4	Monthly	Snow accumulation
			Biweekly	Snowmelt
Evaporation	Class A pan	1	Daily	Nonfreezing periods
	Meteorological raft	1	Continuous	Nonfreezing periods
Ground water	Piezometer	1	Continuous	Year-round
	Piezometers (single plus nests)	34(R)[1] 46(E)[1]	Monthly	Year-round
	Lake-stage gages	7(R) 4(E)	Monthly	Nonfreezing periods

[1] R = Round Lake; E = East Eightmile Lake.

Table 9-2. Description of instrumentation for water-quality mass-balance data collection

Mass-balance component	Sampling equipment	Number of samples per watershed	Frequency of sampling	Period of sampling
Lake storage	Kammerer sampler[1]	4	Monthly	Year-round
Precipitation	Bulk collectors[2]	3	Biweekly to monthly	Year-round
	Wetfall-dryfall collector	1(R)[3]	Daily (wetfall) Biweekly (dryfall)	Year-round
	Adirondack snow tube (snow courses)	4	Biweekly	Snowmelt
Ground water	Gas-squeeze pump (PVC piezometers)	4(R) 5(E)[3]	Monthly	Year-round

[1]See Chapter 5, p.

[2]See Chapter 3, p. 10-13

[3]R = Round Lake; E = East Eightmile Lake.

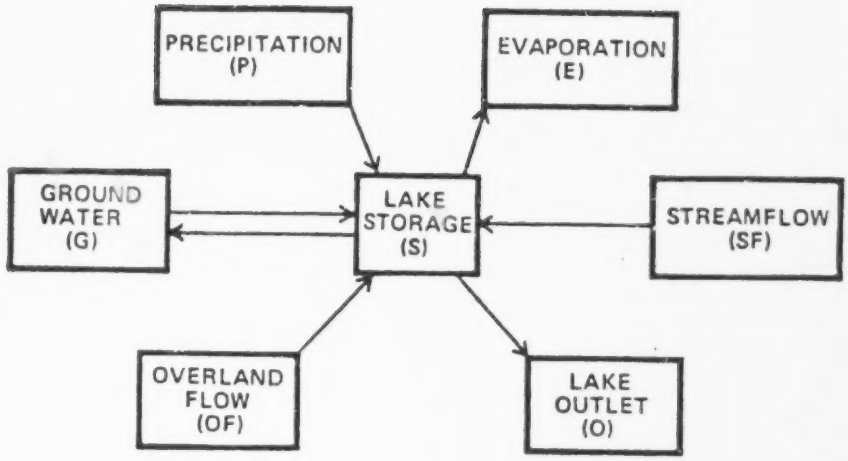


Figure 9-7. Interrelationships among hydrologic-budget components of a typical lake.

Lake Storage - The stage at Round Lake has fluctuated at total of 0.23 m (0.74 ft) during the period July 22, 1981, to November 8, 1982 (Figure 9-8). The maximum monthly fluctuation during this period was 0.10 m (0.32 ft) in May 1982, in response to 119 mm (4.67 in.) of rain that fell in a 16-day period from May 3 through May 18. The minimum monthly fluctuation of 0.01 m (0.04 ft) occurred in December 1981. There has been a slight increase in the mean stage during the period of record.

At East Eight Mile Lake, the maximum fluctuation from July 24, 1981, to November 9, 1982, was 0.28 m (0.92 ft) (Figure 9-9). In July 1982, 276 mm (10.88 in.) of rain caused the maximum monthly fluctuation of 0.19 m (0.62 ft) for the period of record. From July 6 through July 10, 195 mm (7.67 in.) of rain caused the stage to rise 0.16 m (0.53 ft). The minimum monthly stage fluctuation was 0.02 m (0.07 ft) in March 1982. A general decrease in stage occurred from July 1981 through June 1982, but a recent recovery has returned the stage to approximately that existing at the beginning of the period of record.

Precipitation - A total of 805 mm (31.70 in.) of precipitation fell at Round Lake during the 1982 water year; 834 mm (32.84 in.) of precipitation fell at East Eight Mile Lake during the same period. The monthly precipitation distributions as measured by the daily gauges at Round and East Eight Mile Lakes from August 1982 through September 1982, are shown in Figure 9-10. Although the pattern of large and small monthly totals is similar at the two lakes, a correlation analysis of these data provides an r^2 of only 0.75. An analysis of daily values for the same period yields an r^2 of 0.22. Thus, there seems to be little relation between the amounts of precipitation falling at the two sites, particularly on a daily basis.

Regression relationships between rainfall catch in the daily and recording precipitation gauges at the East Eight Mile Lake study area during 1981 and 1982 were very similar (Table 9-3), whereas the relationships for the Round Lake study area during these 2 years appear to be slightly different. Although the equations for 1981 and 1982 have not yet been tested statistically for either study area, the apparent difference at Round Lake may result from the different locations of the daily gauge during the 2 years.

Snow accumulation began in December 1981 at both study areas, peaked at about 500 mm (20 in.) during early February, and was gone by early April. Water equivalent reached 100 to 125 mm (4 to 5 in.) in the Round Lake watershed during mid-March; peaks of 90-115 mm (3.5 to 4.5 in.) were recorded in the East Eight Mile Lake watershed at the same time.

Apparent differences in water equivalent exist among the different cover types in both watersheds. Although these have not been analyzed thoroughly, it is particularly interesting to note that a maximum of about 50 mm (2 in.) water equivalent was found on the ice at Round Lake in early February. A maximum of 25 mm (1 in.) water equivalent on the ice at East Eight Mile Lake was found at the same time. Apparently, the open exposure of the ice surface allows more ablation of the snow cover, particularly by sublimation during the colder months. Also, the wind probably blows snow from the ice.

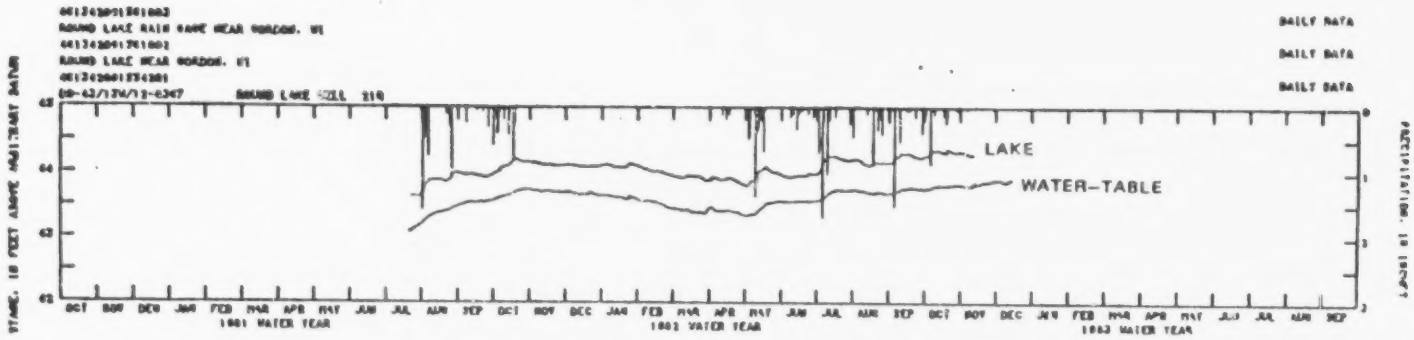


Figure 9-8. Daily rainfall, lake stage, and water-table altitude at the Round Lake study area.

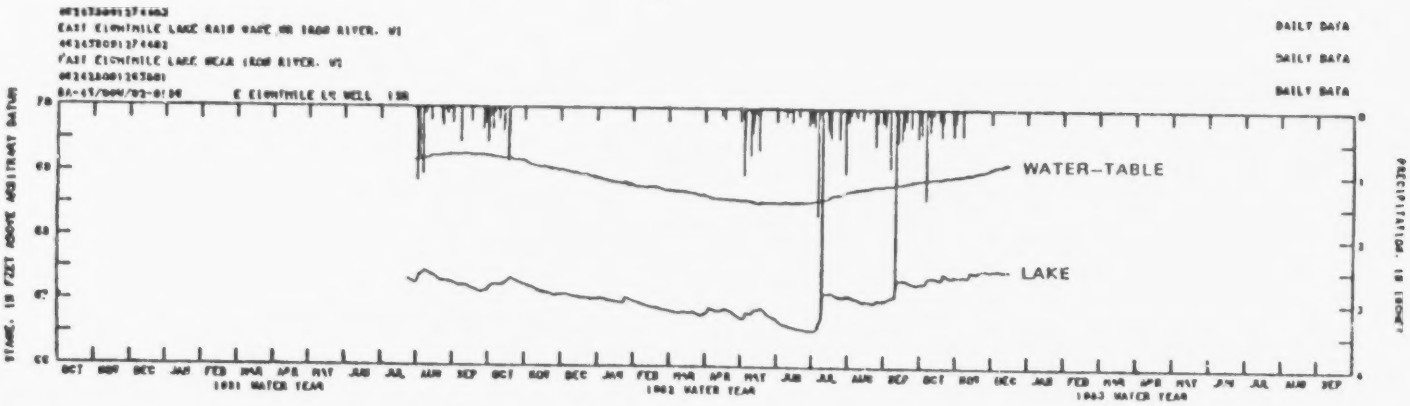


Figure 9-9. Daily rainfall, lake stage, and water-table altitude at the East Eightmile Lake study area.

INTEN

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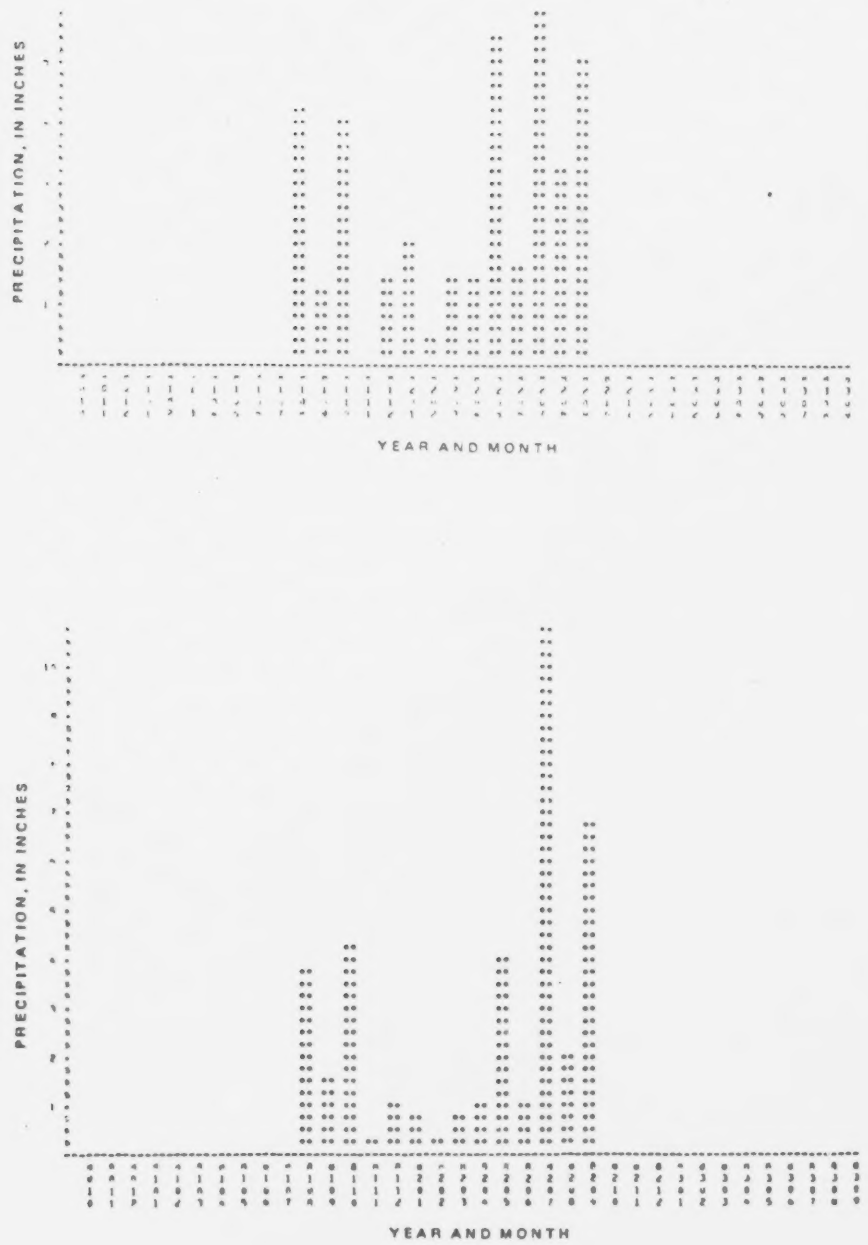


Figure 9-10. Monthly precipitation at Round and East Eightmile Lakes.

Table 9-3. Correlation between Round and East Eightmile Lakes for daily values of meteorological parameters (through September 30, 1982)

Parameter	Squared correlation coefficient (r^2)	Number of observations (n)
Wind speed	0.52	128
Minimum air temperature	.96	261
Maximum air temperature	.98	272
Mean air temperature	.97	246
Relative humidity	.65	183
Vapor pressure	.94	141
Lake-surface temperature	.95	178

Evaporation - Figure 9-11 presents monthly pan-evaporation data for Round and East Eight Mile Lakes for August through October 1981 and for May through September 1982. Pan evaporation at the two study area is similar--587 mm (23.1 in.) at Round Lake from May through September 1982, and 574 mm (22.6 in.) at East Eight Mile Lake for the same period. Correlation between the two areas is better than for precipitation. The value of r^2 for correlation of daily pan evaporation is 0.49; for monthly totals, the value of r^2 is 0.96.

The meteorological parameters measured aboard the rafts generally show good correlation between the two lakes (Table 9-4). This provides a useful procedure for estimating daily values of parameters at one lake from data gathered at the other lake, for periods of instrument failure.

Groundwater - Groundwater levels have fluctuated similarly at the two study areas from July 1981 to November 1982. At Round Lake, fluctuations in shallow piezometers ranged from 0.16 to 0.80 m (0.53 to 2.64 ft) during this period; deep piezometers showed fluctuations of 0.21 to 0.40 m (0.70 to 1.31 ft). Fluctuations in the shallow piezometers at East Eight Mile Lake ranged from 0.17 to 0.63 m (0.57 to 2.06 ft), whereas the deep piezometers registered changes of 0.21 to 0.38 m (0.68 to 1.26 ft). Typical groundwater hydrographs for the two study areas are shown in Figures 9-12 and 9-13.

Figures 9-8 and 9-9 show that groundwater levels at the Round Lake study area seem to respond more rapidly to recharge events. This occurs even though the water table is 4.0 m (13 ft) below land surface at the site of the recording piezometer at East Eight Mile Lake and 11.6 m (38 ft) below land surface at the Round Lake site.

Hydraulic conductivities calculated from slug tests at the two study areas are summarized in Table 9-5. The data indicate that the conductivities at the two areas are very similar, are reasonably uniform, and are typical of clean sands (Freeze and Cherry, 1979).

Figures 9-14 and 9-15 are typical water table contour maps for the study areas. The arrows depicting lateral flow show that both lakes are groundwater flow-through types. East Eight Mile Lake appears to be located in a regional flow system with groundwater entering the lake from the southeast and leaving the lake to the northwest. Round Lake is somewhat more complex. The flow system here seems to be more localized. Groundwater enters the lake from the north and southeast and leaves to the northeast and southwest.

After the hydrologic budgets have been determined, the water quality mass balances for the lakes can be readily calculated knowing the concentrations of chemical constituents in the components of equation 3. And, because lake evaporation is considered to be pure water, this equation can be simplified to:

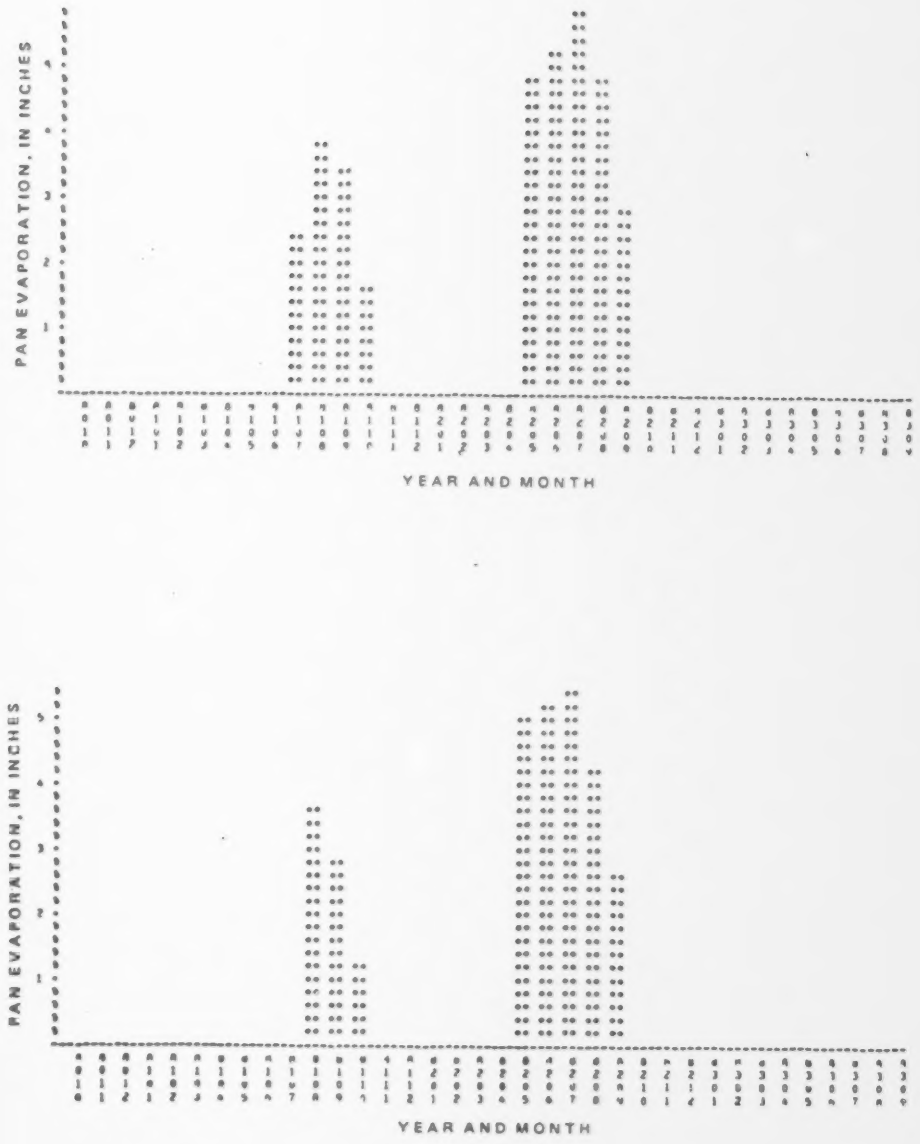


Figure 9-11. Monthly pan evaporation at Round and East Eightmile Lakes.

Table 9-4. Regression relationships between rainfall catch (in millimeters) in daily and recording rain gages

[Daily = (recording X b) + a]

Study area	Period	Regression constant (a)	Regression coefficient (b)	Squared correlation coefficient (r^2)	Number of observations (n)	Approximate range of values (mm)
Round Lake	July 30-Oct. 20, 1981	0.33	0.970	0.960	45	0-40
	May 6-Sept. 30, 1982	.41	1.10	.940	91	0-40
East Eightmile Lake	Aug. 5- Oct. 20, 1981	-.43	.977	.932	46	0-25
	May 11-Sept. 30, 1982	-.001	.967	.836	81	0-75

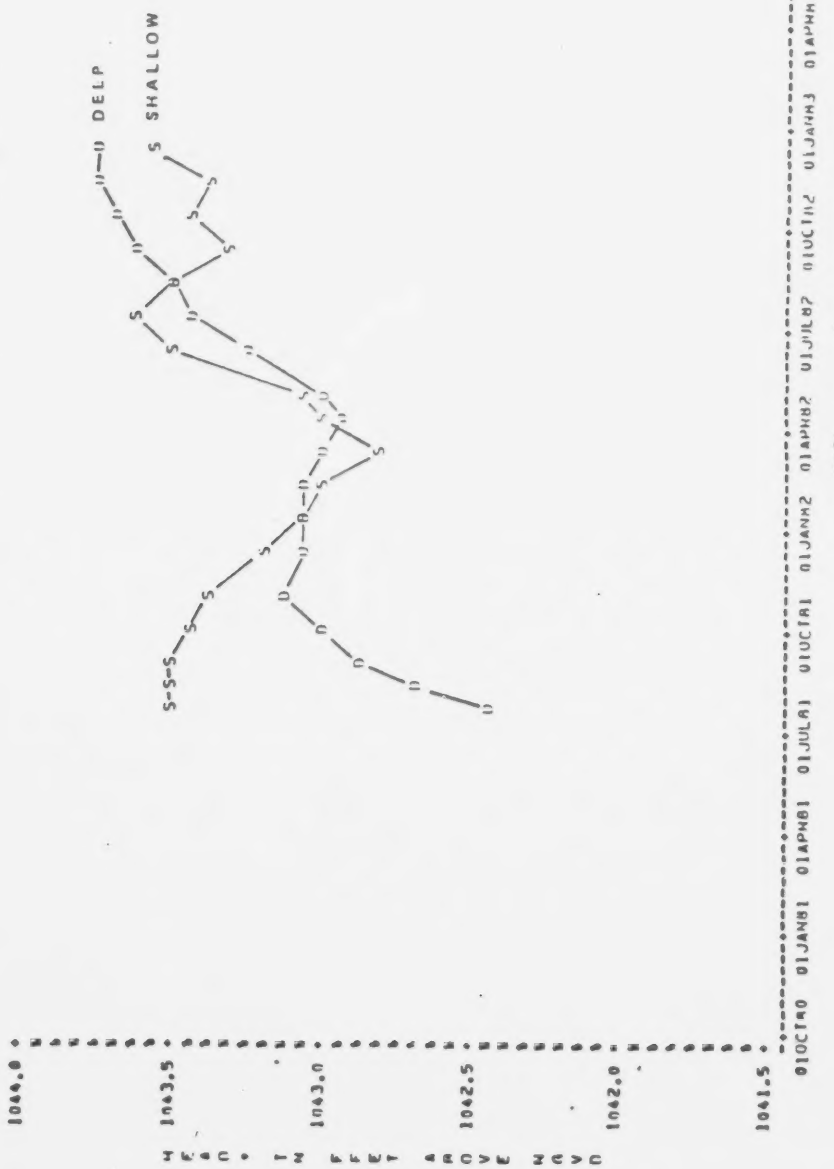


Figure 9-12. Ground-water heads at nest RL12, Round Lake study area.

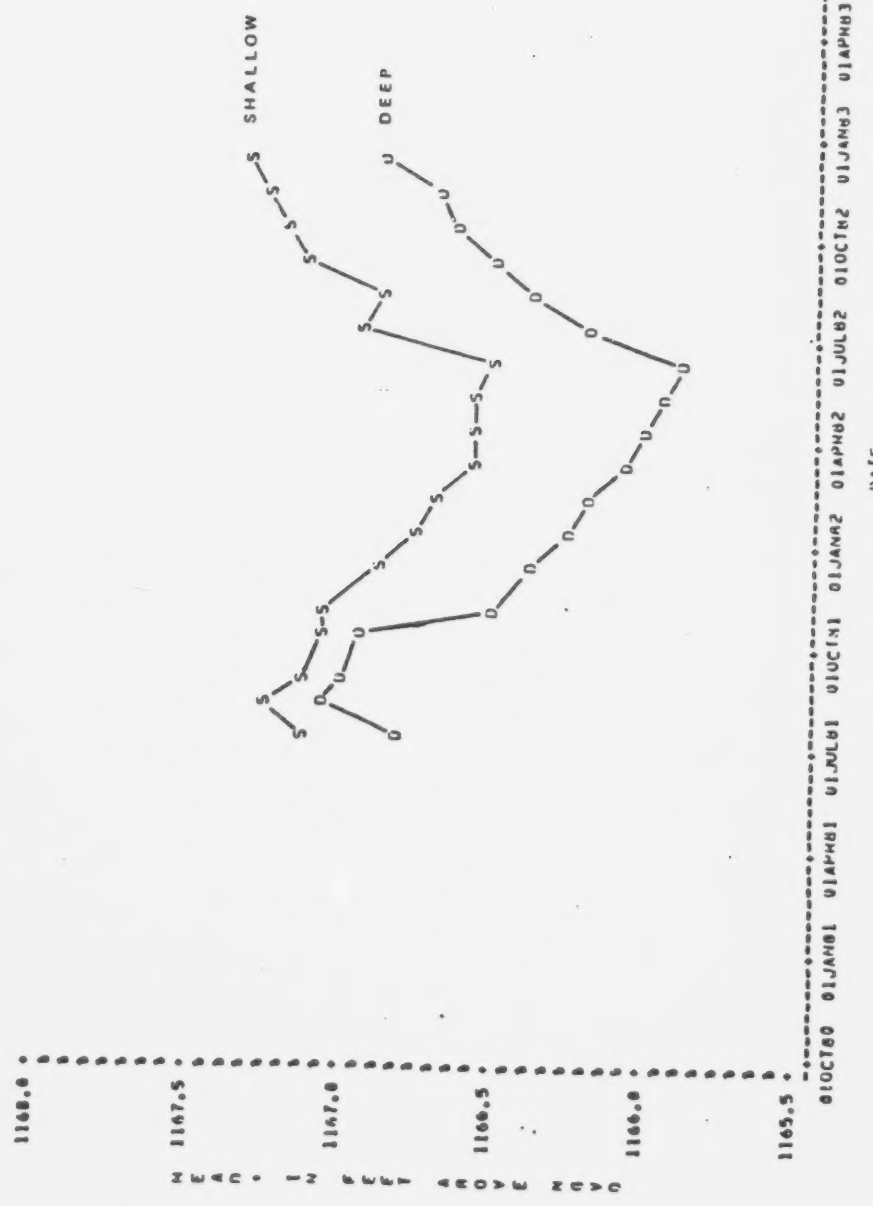


Figure 9-13 Ground-water heads at nest HE07, East Eightmile Lake study area.

Table 9-5. Hydraulic conductivity calculated from slug tests

Study area	Piezometer type	Hydraulic conductivity (m/d)		
		Mean	Standard deviation	Number of observations
Round Lake	Permanent[1]	131	93.8	17
	Temporary[2]	147	72.0	7
East Eightmile Lake	Permanent	178	83.6	14
	Temporary	371	219	6

[1]Surrounding lake.

[2]Lake bottom.

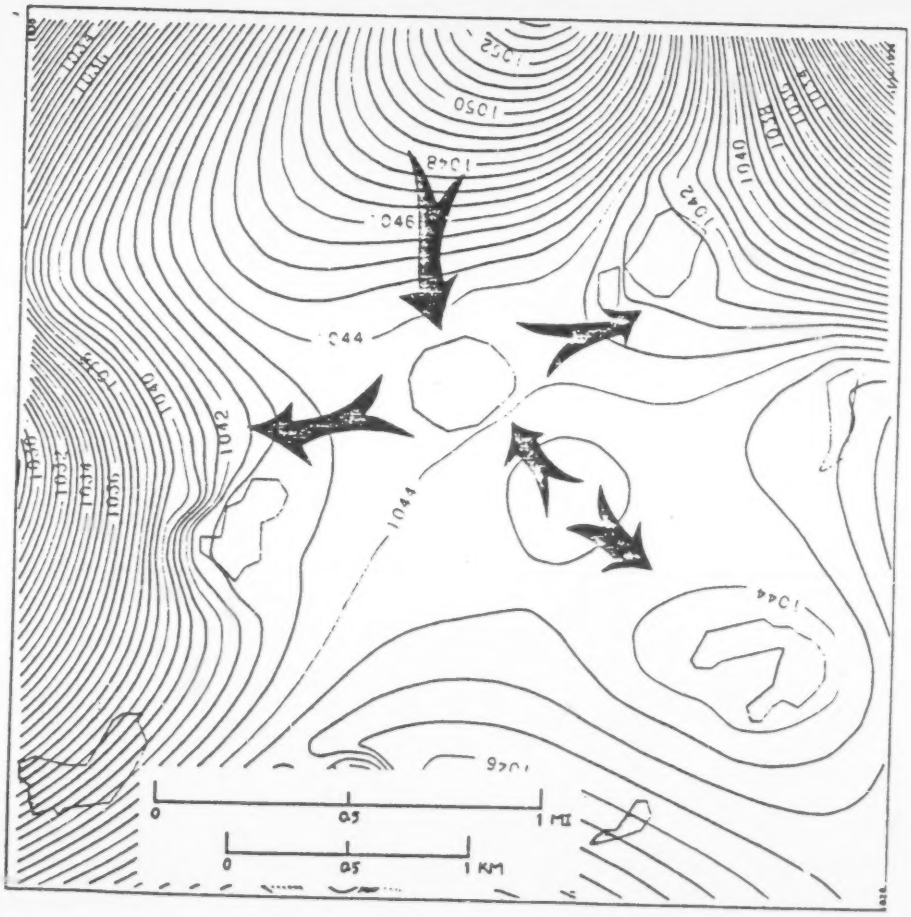


Figure 9-14. Water-table contours (ft above mean sea level) and direction of ground-water flow at Round Lake, August 11, 1981.

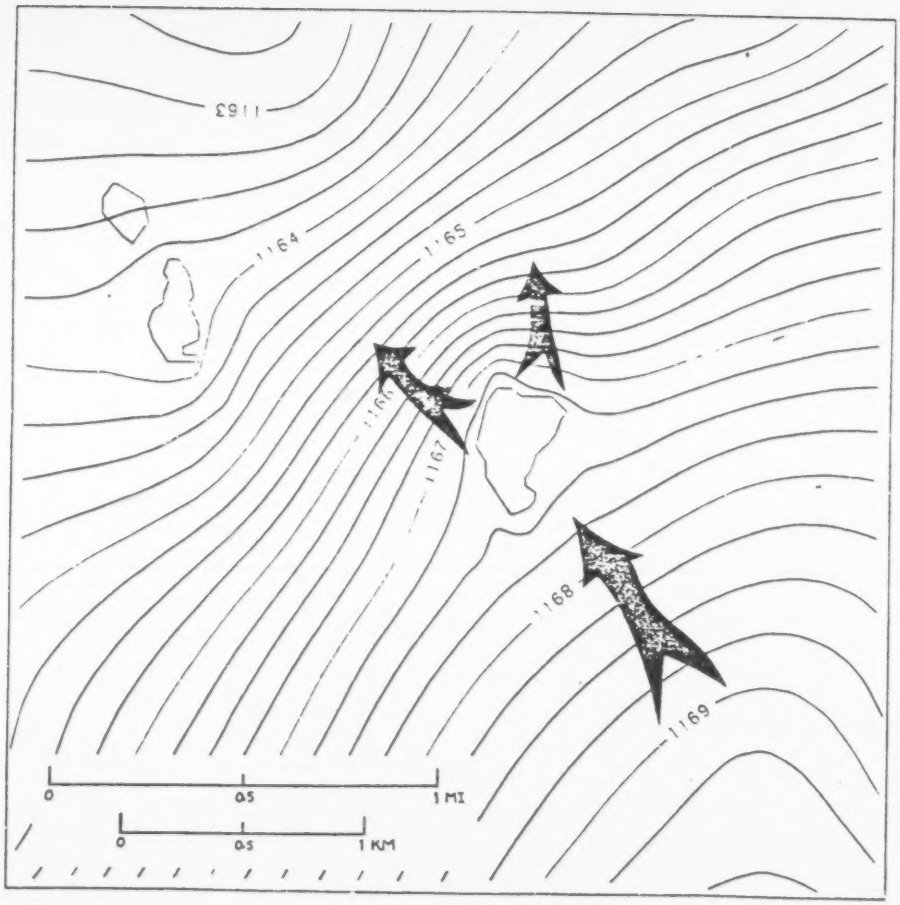


Figure 9-15. Water-table contours (ft above mean sea level) and direction of ground-water flow at East Eightmile Lake, August 12, 1981.

$$S_M = P_M \pm G_M \pm e_M \quad (4)$$

where

S_M is the mass of chemical in storage in the lake,

P_M is the mass of chemical in precipitation on the lake surface,

G_M is the mass of chemical entering or leaving the lake via groundwater flow, and

e_M is the error team.

Snow-course samples were collected on three dates during snowmelt in March 1982. Water equivalent of snow and selected water quality analyses from these samples are shown on Table 9-6.

Comparing the data from the watersheds at each lake, one can see a slight decrease in pH (increase in hydrogen-ion concentration) from March 2 to March 17; from March 17 to March 30, pH increased (hydrogen ion decreased) to values greater than on March 2. Conversely, specific conductance, NH_4 nitrogen, and NO_2 + NO_3 nitrogen increased slightly from March 2 to March 17, whereas by March 30, these parameters had decreased to values less than those existing on March 2.

The change in water equivalent of snow shows that snow accumulated in both watersheds from March 2 to March 17. The increased concentrations of chemical constituents presumably are associated with snow that fell during this period. From March 17 to March 30, snowmelt occurred in both watersheds, and concentrations of chemical constituents decreased in the snow remaining. Apparently, the lost chemical constituents were released with the melt water. If one assumes that specific conductance changes linearly with dilution or concentration by pure water over the small ranges encountered in Table 9-6, and one does not correct for additions from precipitation falling during the period March 17 to March 30 (11.7 mm fell at Round Lake, and 6.9 mm fell at East Eight Mile Lake), then 63 percent of the ion load of the snow at Round Lake was released with 38 percent of the melt water. Similarly, at East Eight Mile Lake, 51 percent of the ion load was released with 26 percent of the melt water. These values compare favorably with data presented by Johannessen and Henriksen (1978), who state that 50 to 80 percent of the chemical load is released with the first 30 percent of the melt water during winters with little or no snowmelt prior to spring.

Changes in chemical composition of snow on lake ice during the spring of 1982 are difficult to interpret, as no snow remained on the ice on March 30. Except for NH_4 nitrogen at Round Lake, changes in pH, specific conductance, NH_4 nitrogen, and NO_2 + NO_3 nitrogen on lake ice from March 2 to March 17 followed patterns similar to those in watershed snow. The changes in concentration, however, were considerably more dramatic. This may be a reflection of the open environment of the lake ice, that is, the lack of interception or modification by a forest canopy.

Table 9-6. Summary of selected water-quality changes during snowmelt, 1982

Study area	Date	Location	Water equivalent of snow (mm)	Field pH (standard units)	Field specific conductance (umhos/cm @25°C)	NH_4 nitrogen (mg N/L)	$NO_2 + NO_3$ nitrogen (mg N/L)
Round Lake	March 2	Watershed snow[1]	105	4.8	14	0.11	0.34
		Lake snow[2]	22	5.1	12	.18	.26
	March 17	Watershed snow	113	4.7	15	.12	.35
		Lake snow	15	4.3	33	.16	.48
		Lake slush		6.0	5	.12	.06
		Lake ice		6.0	4	.04	<.02
	March 30	Watershed snow	70	5.0	9	.10	.18
		Lake snow	[3]	[3]	[3]	[3]	[3]
East Eightmile Lake	March 2	Watershed snow	104	4.8	15	.12	.40
		Lake snow	28	6.0	9	.16	.28
	March 17	Watershed snow	106	4.7	18	.13	.42
		Lake snow	8.9	4.2	42	.21	.62
		Lake slush		7.2	22	.05	.07
		Lake ice		6.2	5	<.02	<.02
	March 30	Watershed snow	78	5.0	12	.11	.25
		Lake snow	[3]	[3]	[3]	[3]	[3]

[1]Water equivalent and chemical concentrations are means of three composite samples (one from each snow course).
[2]Water equivalent and chemical concentrations are for one sample.
[3]No snow present on the ice.

Samples of slush on the ice surface and of lake ice were collected on March 17 for chemical analysis. At Round Lake, chemical composition of the slush was similar to that of the lake ice (Table 9-6). Both had larger pH's and smaller values of specific conductance that the overlying snow. Slush and lake ice at East Eight Mile Lake had different values for pH and specific conductance (Table 9-6), but both had larger pH's and smaller values of specific conductance than the overlying snow.

Specific conductance of snow on the ground at the two study areas is directly proportional to hydrogen-ion concentration. The regression relationship for the field data (excluding slush and lake ice) is

$$K_{SC} = (0.564 \times [H+]) + 5.09, \quad (5)$$

where K_{SC} is specific conductance in umhos/cm at 25°C, and

[H+] is hydrogen-ion concentration, in ueq/L.

This equation explains 95.9 percent of the variation in specific conductance.

Groundwater - Figure 9-16 summarizes selected water quality characteristics for samples from shallow (water table) and deep piezometers at the two study areas. The period covered is from August 1981 to October 1982. Average depth below the water table for the deep piezometers is 29.5 m (97 ft) at Round Lake and 28.4 m (93 ft) at East Eight Mile Lake.

It is apparent that the deep groundwaters from the two study areas are reasonably similar, particularly with regard to major water quality characteristics. On the other hand, the deep groundwater is considerably different from the shallow groundwater within a given study area. This distinction appears more obvious at Round Lake, where the shallow groundwater generally reflects smaller average values for most water quality characteristics than at East Eight Mile Lake.

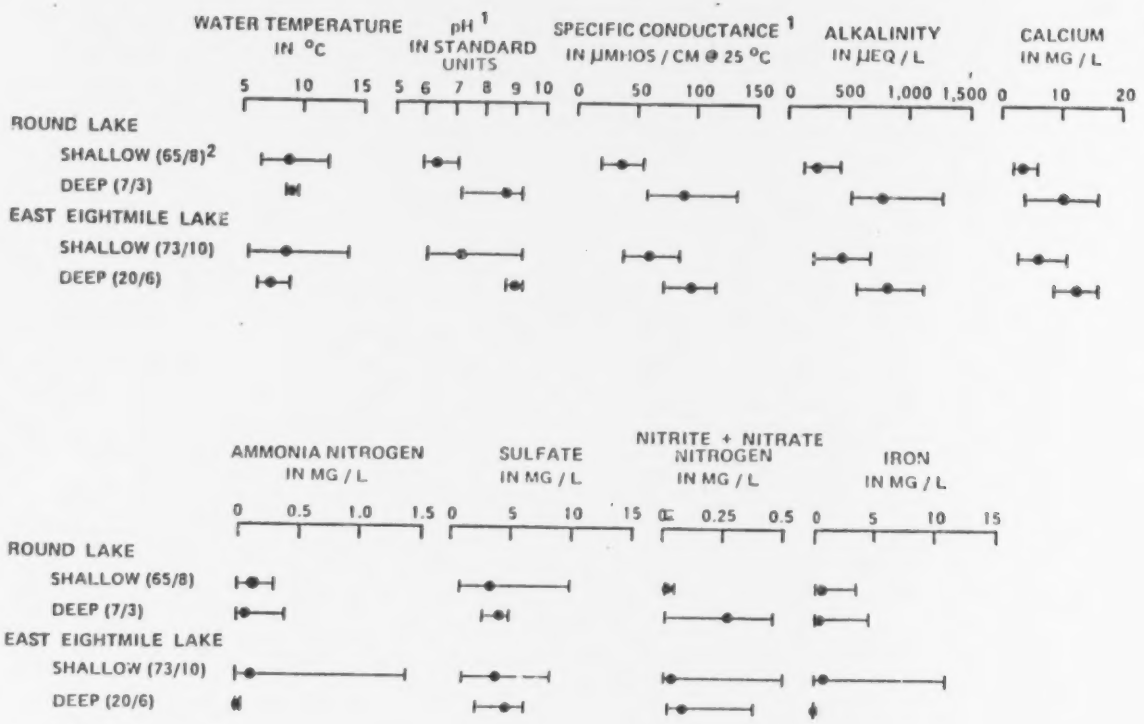
The above conclusions are emphasized by Figure 9-17, which also shows the dominance by Ca and alkalinity among the cations and anions, respectively. The order of decreasing equivalent concentration for cations in shallow and deep groundwater from both study areas is

$$Ca > Mg > (Na+K).$$

For anions, the order of decreasing equivalent concentrations is

$$\text{alkalinity} > SO_4 > (Cl+F+NO_3).$$

Equivalent concentrations of NH_4+Al+Mn+Fe are somewhat tentative because of assumptions regarding the species existing in the groundwater. These data are presented to show that they may be significant components.



[1]FIELD MEASUREMENTS

[2]NUMER OF SAMPLES / NUMBER OF PIEZOMETERS SAMPLED

Figure 9-16. Mean and range of selected water-quality characteristics in samples from shallow and deep piezometers. August 1981 to October 198

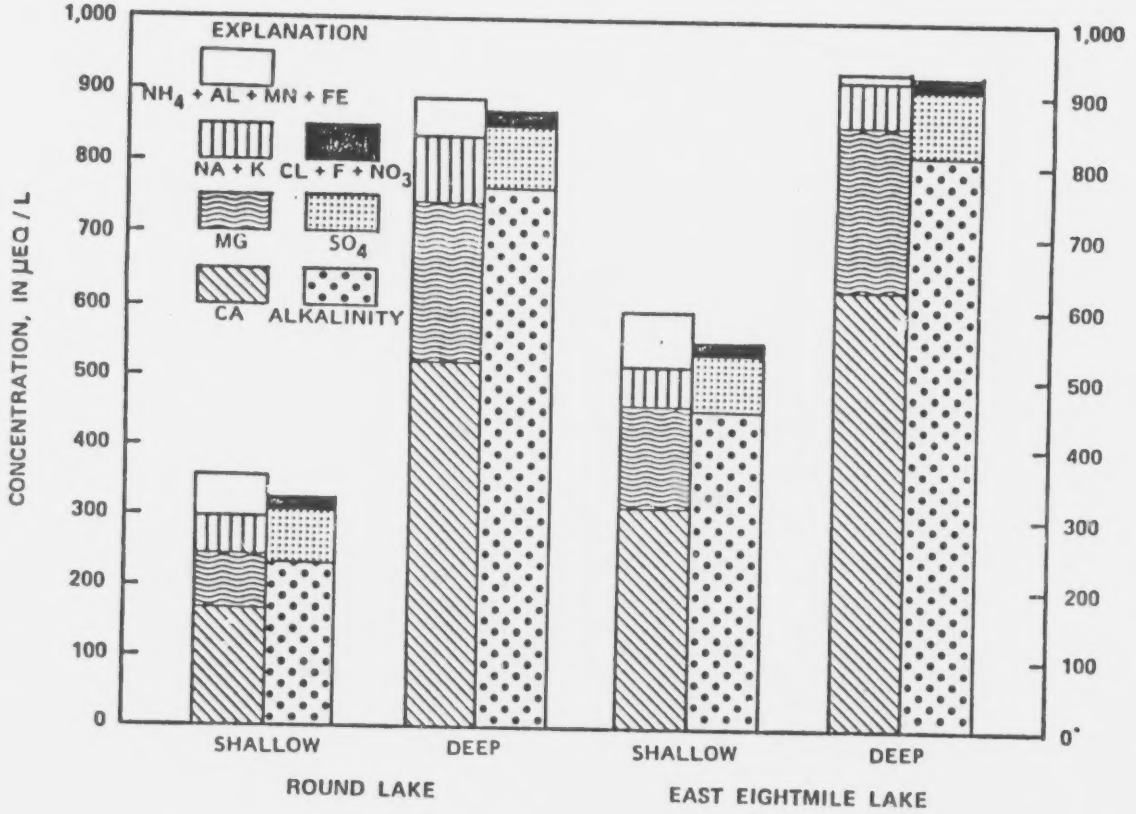


Figure 9-17. Average composition of major cations and anions in samples from shallow and deep piezometers, August 1981 to October 1982.

The primary reason for the generally larger total concentration of chemical constituents in the deep groundwater is the larger concentrations of Ca, Mg, and alkalinity (Figure 9-17). Na+K concentrations are somewhat higher in the deep groundwater at Round Lake. Concentrations of SO_4 and $Cl+F+NO_3$ also are similar in shallow and deep groundwater at both study areas.

Figure 9-18 shows that Ca and alkalinity make up a greater percentage of the major cations and anions in the deep groundwater relative to the shallow groundwater at both study areas.

Chemical analyses for the period August 1981 to June 1982 have been processed using WATEQF (Plummer, Jones, and Truesdall, 1976) to determine mineral phases that may be controlling solution chemistry of the groundwater. Saturation with respect to various mineral phases was evaluated using the ratio of the ion activity product to the thermodynamic equilibrium constant, IAP/KT, where KT indicates the equilibrium constant has been corrected for temperature. If the logarithm of this ratio is positive, then the solution is supersaturated with respect to the mineral phase, if the logarithm is negative, under-saturation is indicated. One of the more interesting results of the analysis relates to the mineral calcite.

At the Round Lake study area, shallow groundwater was considerably under-saturated with respect to calcite; values of log (IAP/KT) ranged from -4.05 to -2.55 for 39 samples from seven piezometers. The deep groundwater generally was at or near saturation; log (IAP/KT) values ranged from -0.24 to 0.04 in six samples from the deep piezometers at RL05 and RL11. One sample from the deep piezometer at RL23 provided a value of -2.29 for log (IAP/KT), indicating under-saturation.

The deep groundwater at the East Eight Mile Lake Study area is at or near saturation with respect to calcite; values of log (IAP/KT) ranged from -0.21 to 0.25 for 14 samples from six piezometers. Except for the area near the south end of the East Eight Mile Lake, the shallow groundwater is considerably under-saturated with respect to calcite. Figure 9-19 shows that the shallow ground water at EE10 is close to saturation; this influence extends to EE11, which is under-saturated, but not nearly so much so the rest of the shallow groundwater. The shallow groundwater at EE10 reached saturation with respect to calcite during the winter of 1981-82 (Figure 9-20). Values of log (IAP/KT) in the shallow groundwater at this site are slightly less than in the deep groundwater; the values appear to be cyclic and follow similar patterns.

Bivariate regression analyses of selected water quality characteristics versus field specific conductance for the combined study areas are summarized in Table 9-7. The data show that Ca, Mg, alkalinity, and dissolved solids correlate well with specific conductance, and reflect the fact that most changes in total ionic composition result from changes in Ca, Mg, and alkalinity.

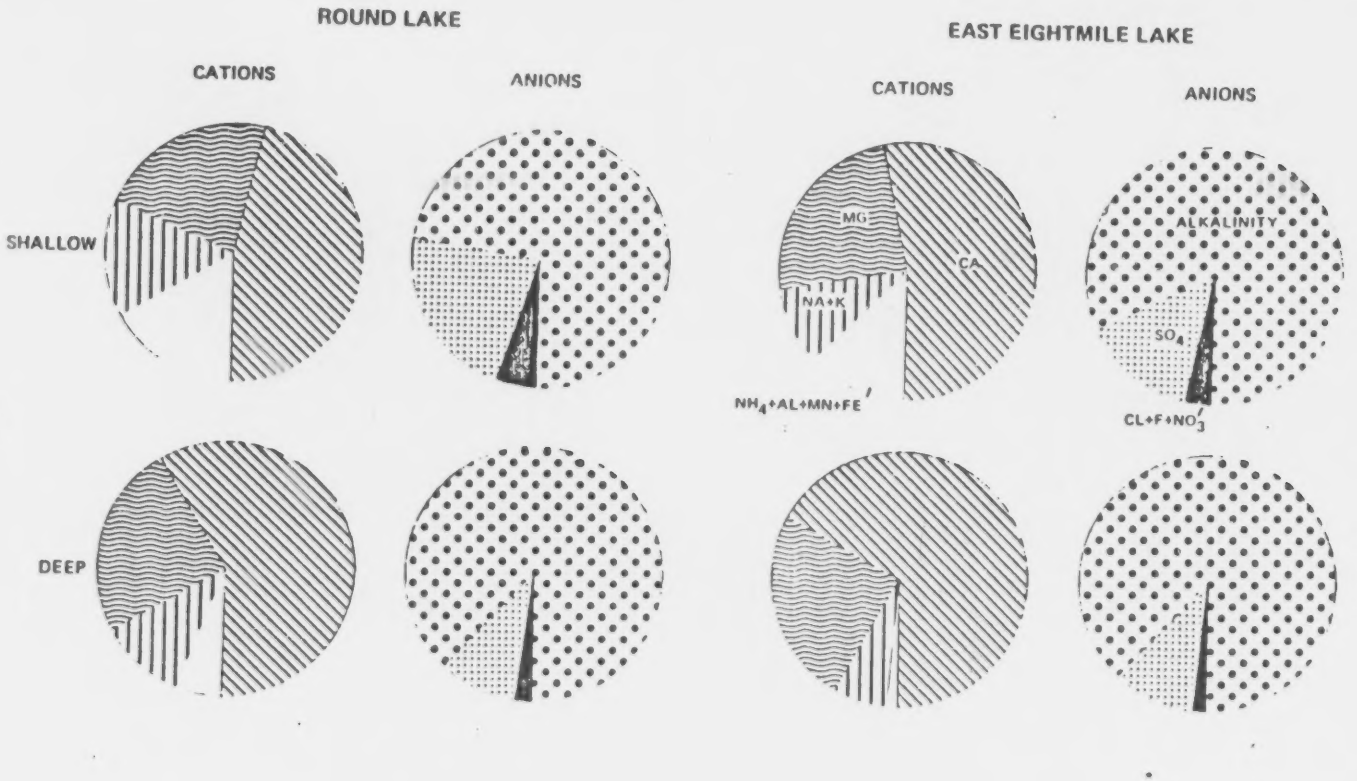


Figure 9-18. Average percentage composition of major cations and anions in samples from shallow and deep piezometers, August 1981 to October 1982.

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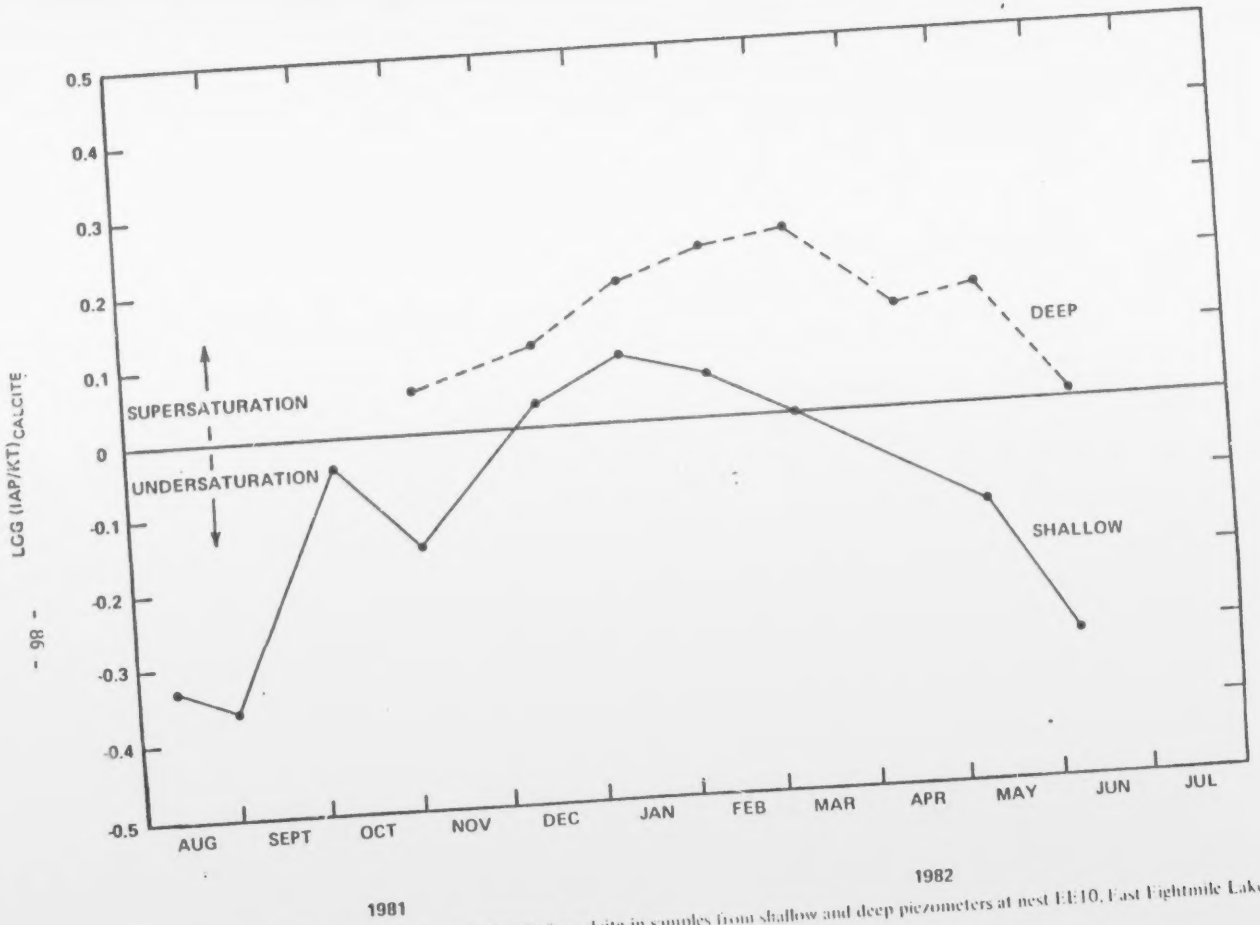

Figure 9-19. Seasonal variations in log (IAP/KT) for calcite in samples from shallow and deep piezometers at nest EE10, East Eightmile Lake.

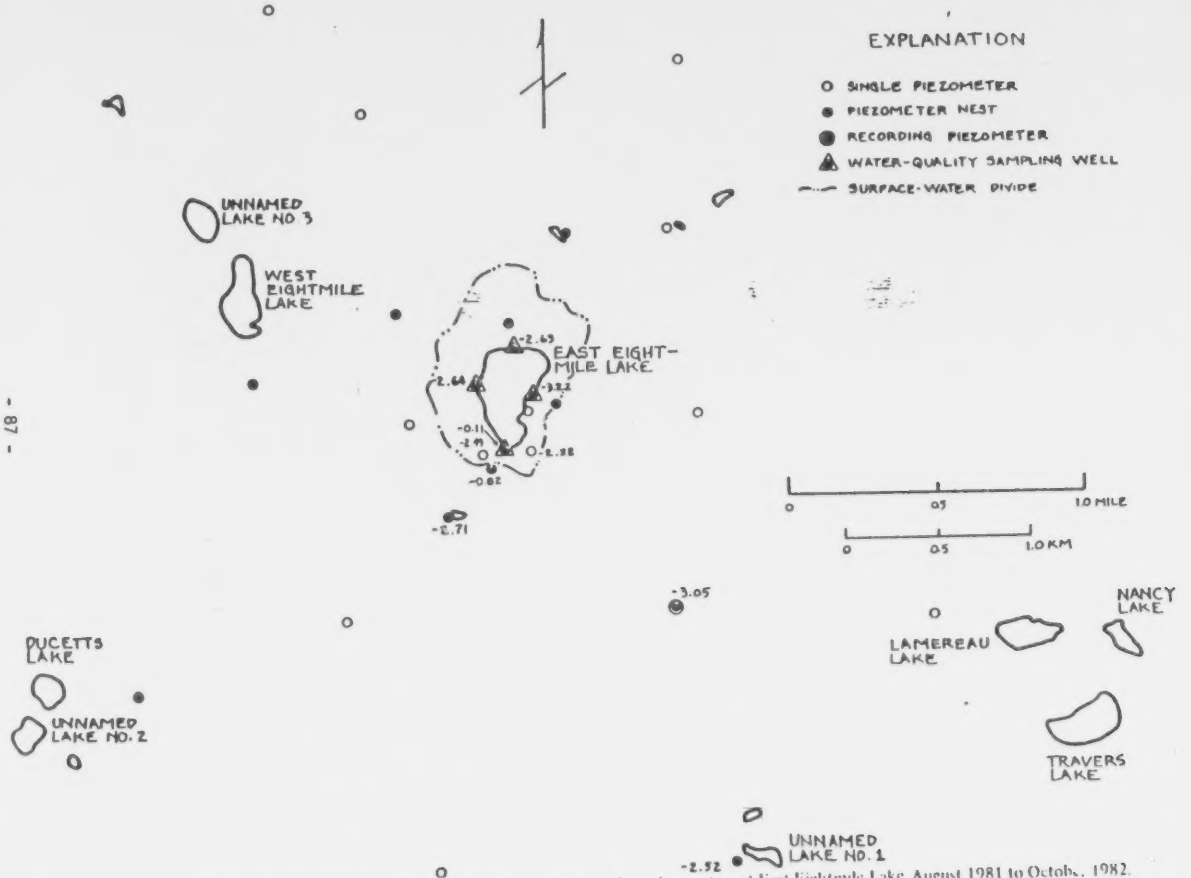

Figure 9-20. Areal variations in log (IAP/KT) for calcite in samples from flow piezometers at East Eightmile Lake, August 1981 to Octob. 1982.

Table 9-7. Regression relationships between selected water-quality characteristics and field conductance for the combined study areas

[Characteristic = (field specific conductance X b) + a]

[Range of specific conductance: 20-133 umhos/cm @ 25°C]

Characteristic	Units	Regression constant (a)	Regression coefficient (b)	Squared correlation coefficient (r^2)	Number of observations (n)
Ca	mg/L	-2.24	0.148	0.929	161
Mg	mg/L	-1.36	.0312	904	159
NH_4-N	mg/L	[1]	[1]	.0839	94
Fe	mg/L	[1]	[1]	.142	96
SO_4	mg/L	[1]	[1]	.0644	93
SiO_4	mg/L	[1]	[1]	.288	161
Field alkalinity	ueq/L	-61.1	.505	.848	164
Calculated dissolved solids	mg/L	.894	.880	.944	150

[1]Not given because of the low values of r^2.

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Routine hydrologic and water quality data collection will continue through September 1983. Two additional intensive field activities are planned. One is a general water quality reconaissance of all wells at both study areas. Field measurements of water temperature, pH, specific conductance, and alkalinity will be made. These data should help determine whether the shallow and deep ground waters represent different flow systems and source areas or, simply, different contact times. The second field activity is a seismic study of the two areas to determine depth to bedrock. This information will allow better definition of the groundwater interaction with the lakes.

Preliminary hydrologic and water quality mass-balance budgets for the two lakes will be completed during 1983.

Limnological Component

Following is a summary of the progress and information gathered on the limnology of the two study lakes.

Although this project did not officially begin until August, 1981, some limnological sampling actually started earlier and is included in this report. Samples were collected from 3 sites in each lake, with the deepest spot in each lake the primary site where profile samples was carried out.

Round Lake is relatively deep and as a result is dimictic. Strong stratification develops during the summer months, and bottom waters become anaerobic both in summer and late winter. East Eight Mile Lake, because of its shallower depth, is polymictic with good dissolved oxygen concentrations throughout the year.

The pH of Round Lake during the reporting period ranged from 5.8-8.9, and averaged 6.5. The wide range in pH appeared to be primarily caused by seasonal changes in phytoplankton productivity. Mean pH in East Eight Mile Lake (6.9) was higher than in Round Lake and the range was narrower (6.3-8.2). Field alkalinity of East Eight Mile Lake averaged 24 mg/l (range 16-41), while the mean Gran alkalinity of Round Lake was 6.6 mg/l (range 4.3-13.0). Average conductivity of Round and East Eight Mile Lake was 25 and 55 umhos/cm, respectively.

Round Lake produced more chlorophyll a than East Eight Mile, indicative of the greater algal biomass, but East Eight Mile had more macrophytes. Mean chlorophyll a concentration in Round Lake was 8 mg/m^3 and the maximum was 30 mg/m^3; the lake experienced blue-green algal blooms of Anabaena affinis in both 1981 and 1982. Peak chlorophyll a level was only 7 mg/m^3 in East Eight Mile Lake and averaged only 3 mg/m^3. Correspondingly, phosphorus concentrations are higher in Round Lake (.032 mg/l) than in East Eight Mile (.010 mg/l).

Heavy metal concentrations were measured continuously in both lakes during the early stages of the study and periodically thereafter, but samples were consistently below laboratory detection limits. Concern has been expressed elsewhere about the mobilization of aluminum as a result of acid precipitation, but none was detected in the study lakes and there is no evidence of toxicity to biota.

A large number of species of phytoplankton and zooplankton inhabit Round and East Eight Mile Lakes, and some of the species present have been found elsewhere to be sensitive to acidification. Thus, the plankton species composition of the study lakes shows no apparent impact from acid precipitation.

East Eight Mile Lake has 8 species of fish and Round Lake 5, not counting introduced species. Round Lake has populations of smallmouth bass and bluntnose minnows, both reported as sensitive to pH below 6.0. The bluntnose minnow population is thriving, but the smallmouth bass population is not reproducing and appears on the verge of extinction. However, factors other than low pH are most likely the cause for the decline of the population; possibilities include competition from other species, lack of spawning substrate, lack of cover, and absence of important food organisms. Changes in fish populations, sometimes drastic, are not unusual in Wisconsin lakes, and these changes are seldom explainable.

The limnological sampling sites for Round and East Eight Mile Lakes are shown in Figures 9-21 and 9-22. For water chemical analyses, samples were collected in the deepest area in both lakes (RL-1 and EL-1). In East Eight Mile Lake samples were also collected at EL-2 and EL-3. Phytoplankton, zooplankton, water clarity, and water temperature information was collected at the deepest water depth as well as at two nearshore stations in both lakes. The water depths at the sampling sites in Round Lake were RL1 - 22.5 meters, RL2 - 4 meters, and RL3 - 4 meters. The sampling sites in East Eight Mile Lake had water depths of EL1 - 6.5 meters, EL2 - 3 meters, and EL3 - 3 meters. The water depths of the four benthic invertebrate sites at Round Lake were RLI - 22.5 meters, RLII - 12 meters, RLIII - 15 meters, and RLIV - 10.5 meters. The water depths of the sampling sites at East Eight Mile Lake were ELI - 6.5 meters, ELII - 3.5 meters, ELIII - 6.5 meters, and ELIV - 4.5 meters.

The temperature isotherms for Round Lake are shown in Figure 9-23. At the beginning of sampling in July 1981 the lake was stratified with the thermocline beginning at 3 meters. The lake mixed completely in November with ice formation occurring about December 6. Ice cover reached depths of about 50 cm overlain by slush and snow depths up to 40 cm. The ice cover broke up on April 23, 1982 at which time the lake was well stratified with the surface temperature exceeding 14°C.

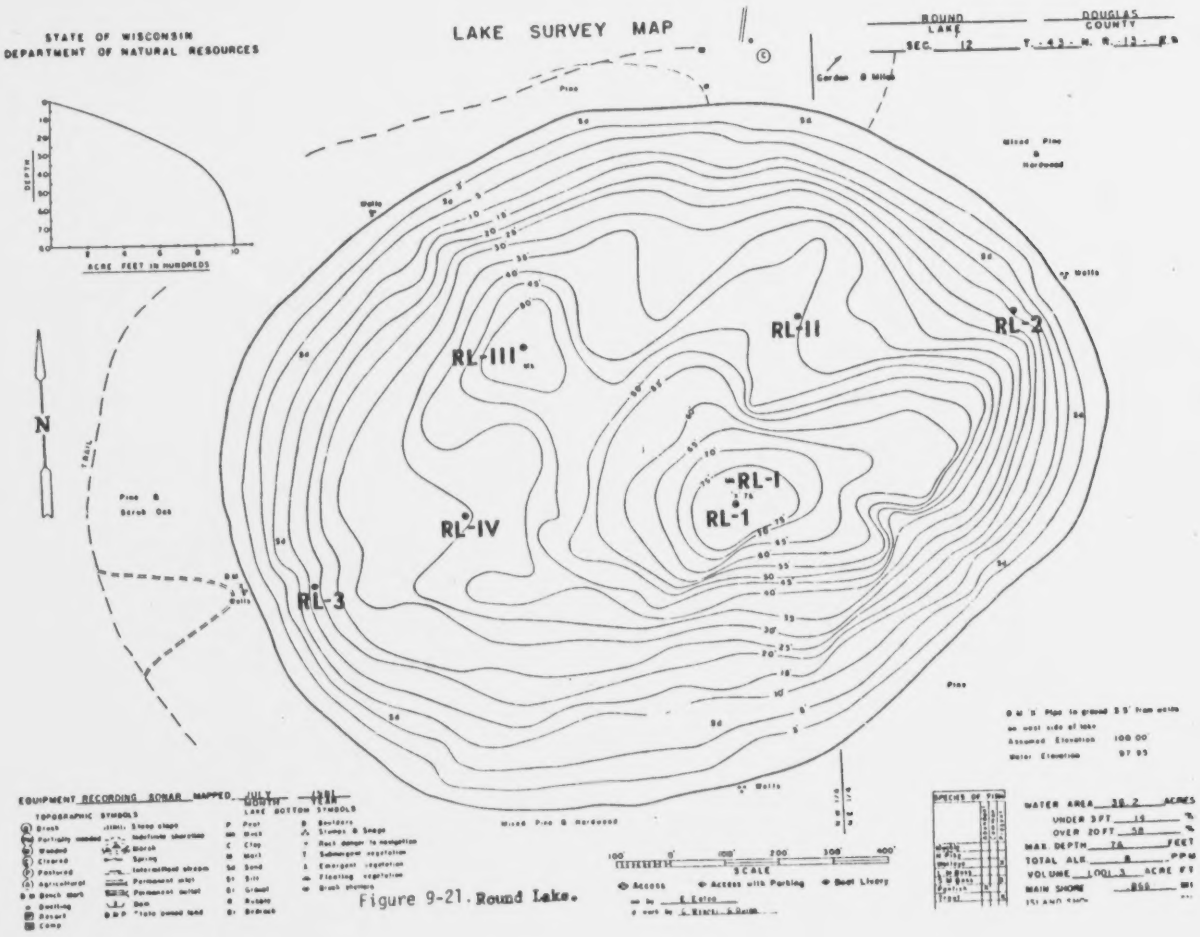


Figure 9-21. **Round Lake.**

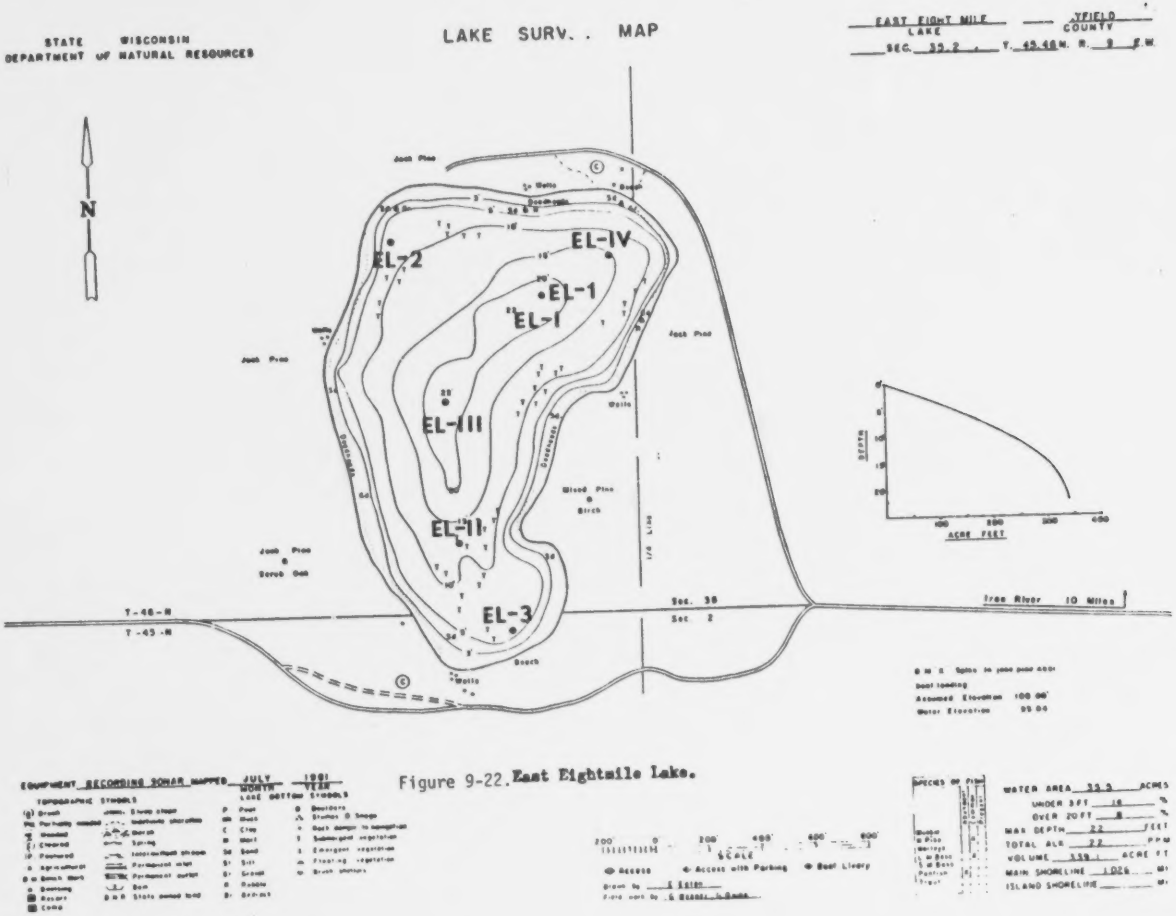


Figure 9-22. East Eightmile Lake.

In contrast to Round Lake, East Eight Mile Lake never exhibited true stratification with formation of a real thermocline and hypolimnion. Occasionally there was some difference in temperature and dissolved oxygen concentration from surface to bottom, but these conditions lasted for only short periods of time (Figure 9-24). Surface temperatures on East Eight Mile Lake were the same as on Round Lake during the summer, and ice formation occurred at about the same time on both lakes. Also, ice thickness was nearly the same and ice-out occurred at about the same time in the spring.

Round Lake exhibited a metalimnetic oxygen maximum during parts of the summer period of stratification in both years (Figure 9-23). Dissolved oxygen concentrations exceeded 13 mg/l in 1981 and reached 13 mg/l in 1982. During summer stagnation the lower portion of the hypolimnion became anaerobic. In 1981 this first occurred in August and reached an upper limit of 16 meters, while in 1982 the anaerobic period began earlier and extended to 13 meters. The bottom waters also became anaerobic during the later part of the ice covered period for about six weeks.

Dissolved oxygen concentrations in East Eight Mile Lake were more consistent (Figure 9-24) primarily because the lake did not develop true stratification. During the ice covered period oxygen levels were somewhat reduced in the bottom waters but concentrations remained high in the upper waters. The only time summer oxygen concentrations were not similar throughout the water column was in late July 1982 when the lake had a temperature difference of 2.5°C from top to bottom; the DO level found in the bottom waters at this time was 2.3 mg/l.

Ranges and arithmetic means of the measured chemical, physical, and biological parameters for both lakes are shown in Tables 9-8 and 9-9. Mean field alkalinity for Round Lake was 9 mg/l, but Gran alkalinity was lower - 6.6 mg/l. Gran alkalinity is considered to represent the true alkalinity of Round Lake, however, field alkalinity is most comparable to historical alkalinity data on Wisconsin lakes because the methods used are the same. In East Eight Mile Lake the mean field alkalinity was 24 mg/l which was similar to the Gran alkalinity value of 23.6 mg/l. Because of the good comparability of the alkalinity values measured in the field by the standard titration method and the values measured in the laboratory by Gran technique, Gran titration of East Eight Mile Lake samples was discontinued.

Field pH values found in Tables 9-8 and 9-9 represent meter readings of nonaerated samples; pH measurements by other methods have been continued, but the field meter values are believed to be the most representative of actual lake pH. Average pH for Round Lake was 6.5 and the range was from 5.8 - 8.9. East Eight Mile Lake had a higher mean pH (6.9) and a narrower range (6.3 to 8.2).

Conductivity was higher in East Eight Mile Lake (55 umhos/cm) than Round Lake (25 umho/cm). Nutrients, especially phosphorus, were higher in Round Lake than East Eight Mile Lake, which was manifested in higher average chlorophyll *a* concentrations in Round Lake. Consistent with what would be

ROUND LAKE

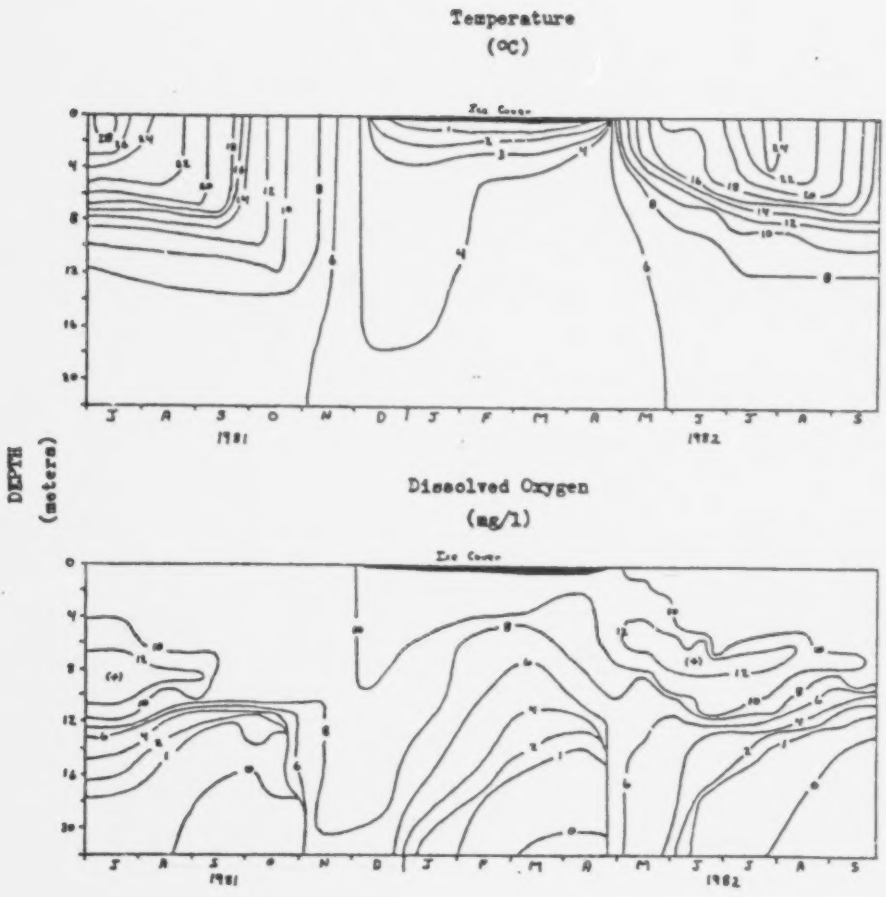


Figure 9-23. **Isotherms and DO isopleths for Round Lake.**

EAST EIGHTMILE LAKE

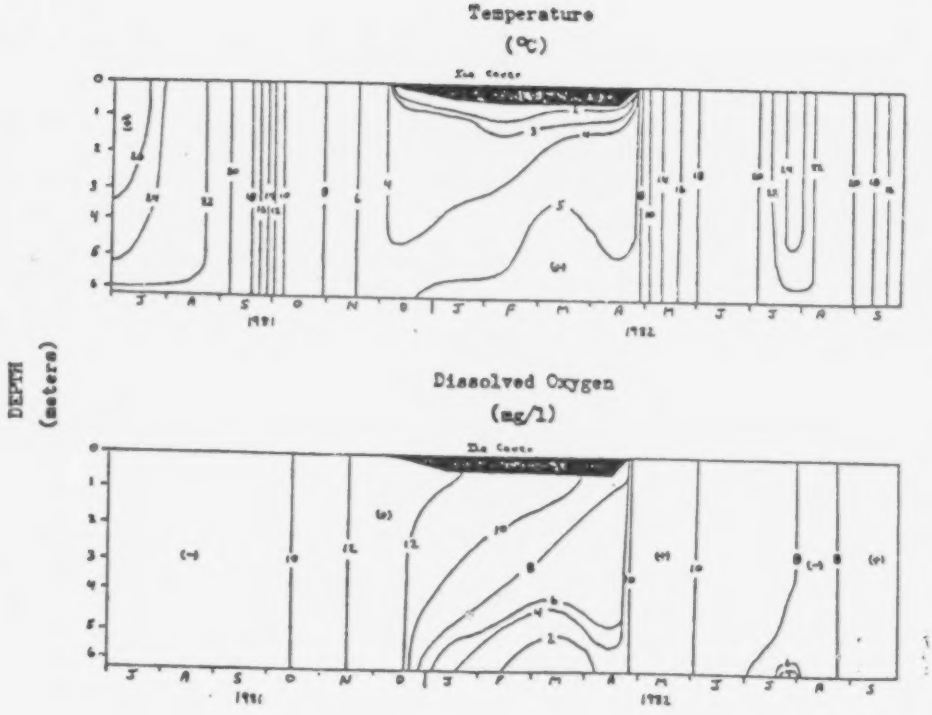


Figure 9-24. Isotherms and DO isopleths for East Eightmile Lake.

Table 9-8. Ranges and Means of Water Quality Data from Round Lake, August 1981 - September 1982.

Parameter	Number of Samples	Range	Mean
Alkalinity (Field (mg/l)	131	6-22	9
(ueq/l)	131	120-440	180
Alkalinity (Lab) (mg/l)	56	4-18	9
(ueq/l)	56	80-360	180
Alkalinity (Gran) (mg/l)	28	4.3-13.0	6.6
(ueq/l)	28	86-260	132
pH (field)	130	5.8-8.9	6.5
pH (lab)	56	6.0-9.1	6.7
Conductivity (field) (umhos/cm)	125	10-58	25
Conductivity (lab) (umhos/cm)	56	18-50	24
Ca (mg/l)	56	1.8-3.0	2.1
Mg (mg/l)	56	0.3-1.0	0.6
K (mg/l)	56	0.4-1.0	0.6
Na (mg/l)	56	0.4-1.0	0.6
Cl (mg/l)	28	.2-0.6	0.3
SO_4 (mg/l)	27	2.0-3.0	2.4
$Si0_2$ (mg/l)	56	.2-1.0	0.3
Total-P (mg/l)	56	0.009-0.174	0.032
DRP (mg/l)	56	.003-0.124	0.012
NH_4-N (mg/l)	56	.02-0.90	0.18
NO_2+NO_3-N (mg/l)	56	.02-0.43	0.09
TKN (mg/l)	56	0.2-1.5	0.5
Fe (mg/l)	56	.1-2.5	0.4
MN (mg/l)	56	.04-0.30	0.07
F (mg/l)	16	.1	.1
Al (mg/l)	40	.025	.025
Cu (mg/l)	16	.003	.003
Pb (mg/l)	16	.003	.003
Zn (mg/l)	16	.020	.020
Hg (mg/l)	16	.0002	.0002
Color (FTU)	118	5-70	8
Chl *a* (mg/m^3)	127	1.2-29.6	8.1
Temp (°C)	424	0.0-24.5	9.8
D.O. (mg/l)	315	0.0-13.2	6.3
Secchi (m)	23	1.5-10.1	5.2

Table 9-9. Ranges and Means of Water Quality Data from East Eightmile Lake, August 1981-September 1982.

Parameter	Number of Samples	Range	Mean
Alkalinity (field) (mg/l)	70	16-41	24
(ueq/l)	70	320-819	474
Alkalinity (lab) (mg/l)	55	20-46	25
(ueq/l)	55	400-919	494
Alkalinity (Gran) (mg/l)	16	18.1-38.5	23.6
(ueq/l)	16	362-769	472
pH (field)	70	6.3-8.2	6.9
pH (lab)	56	6.8-9.0	7.4
conductivity (field) (umhos/cm)	56	35-96	55
conductivity (field) (umhos/cm)	56	42-101	54
Ca (mg/l)	56	5-15	6.3
Mg (mg/l)	56	1.4-2.1	1.8
K (mg/l)	56	0.1-1.0	0.3
Na (mg/l)	56	0.8-1.2	1.0
Cl (mg/l)	24	0.3-0.8	0.4
SO_4 (mg/l)	28	2.6-3.8	3.1
SiO_2 (mg/l)	56	.2-1.4	0.6
Total-P (mg/l)	56	0.005-0.033	0.010
DRP (mg/l)	56	.004-0.013	.004
NH_4-N (mg/l)	56	.02-0.55	0.05
NO_2+NO_3-N (mg/l)	56	.02-0.17	0.04
TKN (mg/l)	56	0.4-1.0	0.5
Fe (mg/l)	40	.1-1.5	.1
Mn (mg/l)	40	.04-0.1	.04
F (mg/l)	16	.1	.1
Al (mg/l)	32	.025-0.033	.025
Cu (mg/l)	16	.003	.003
Pb (mg/l)	16	.003	.003
Zn (mg/l)	16	.020	.020
Hg (mg/l)	20	.0002	.0002
Color (FTU)	60	5-10	5
Chl *a* (mg/m^3)	68	0.7-6.9	2.7
Temp (°C)	124	0.0-24.5	14.0
DO (mg/l)	106	2.0-12.8	9.0
Secchi (m)	23	4.0- 6.7	6.0

expected, water transparency as measured by Secchi disc was less in Round Lake than in East Eight Mile. Mean Secchi depth would be even greater than it was in East Eight Mile Lake if the lake were deeper since many time the Secchi disc was still visible when it was on the lake bottom. As reflected by the conductivity values, most of the major cations and anions are present in higher concentration in East Eight Mile Lake. This is especially true of calcium and bicarbonate and to a lesser extent magnesium. The waters of both lakes can be classified as calcium carbonate waters. The only heavy metals present in measurable quantities were iron and manganese. Aluminum, which has reported as a possible problem in lakes impacted by acidic precipitation, was never detectable (detection limit - 0.025 mg/l) in Round Lake and only once in East Eight Mile Lake (which could have been a sampling or laboratory error).

Seasonal trends of pH, alkalinity, and conductivity for Round Lake are shown in Figure 9-25. The lowest pH value measured was in July 1981, before the official start of the project. By early September the pH had increased to 7.1. From early September on through fall, 1981, pH steadily declined until it reached a plateau of 6.2-6.4 in November and remained at that level through the ice covered period. With the exception of the July 1981 sample the minimum pH values were recorded during spring thaw in early April, 1982. The spring decline was not great - the minimum pH value recorded was 6.0, which was similar to levels in November 1981. Following the melting of the ice cover, pH increased and reached a maximum value of 8.9 in late July. These high pH's were measured in the afternoon (1500 hours) during an algal bloom; high pH occurred not only at the surface but throughout the epilimnion.

As was the case with pH, the minimum field alkalinity was measured in July 1981. Beginning in August the alkalinity increased until late September when it reached 10 mg/l, where it stayed throughout the period of ice cover. After ice-out, the alkalinity decreased to 7 mg/l in May, 1982, and remained there through September 1982. Alkalinity values determined by the Gran technique were consistently lower in Round Lake than those measured by titration to a color change endpoint. Dillon et al. (1978) found that in very dilute waters of south-central Ontario alkalinities measured to a pH endpoint of 4.5 overestimated the buffering capacity by 17 to 48%; they found that the pH endpoint actually varied between 5.0 and 5.5. Alkalinity values measured by the Gran titration method from March through May were consistently 4.9 - 5.4 mg/l, but increased somewhat during the rest of the sampling period to 6.8 mg/l in September. During the summer stratified period in both years alkalinity was generally lower in the epilimnion than the hypolimnion. The differences were greatest in August when surface values were 5.2 mg/l while hypolimnetic values were 11.1 mg/l.

Figure 9-26 shows the seasonal trends of pH, alkalinity, and conductivity for East Eight Mile Lake. With the exception of early September, pH values were generally about the same (6.6-6.8) from July 1981 through mid-April, 1982. Field pH was generally higher than in Round Lake during this period. No pH depression was experienced in East Eight Mile Lake during spring melt. Following spring break-up pH steadily increased to a

ROUND LAKE

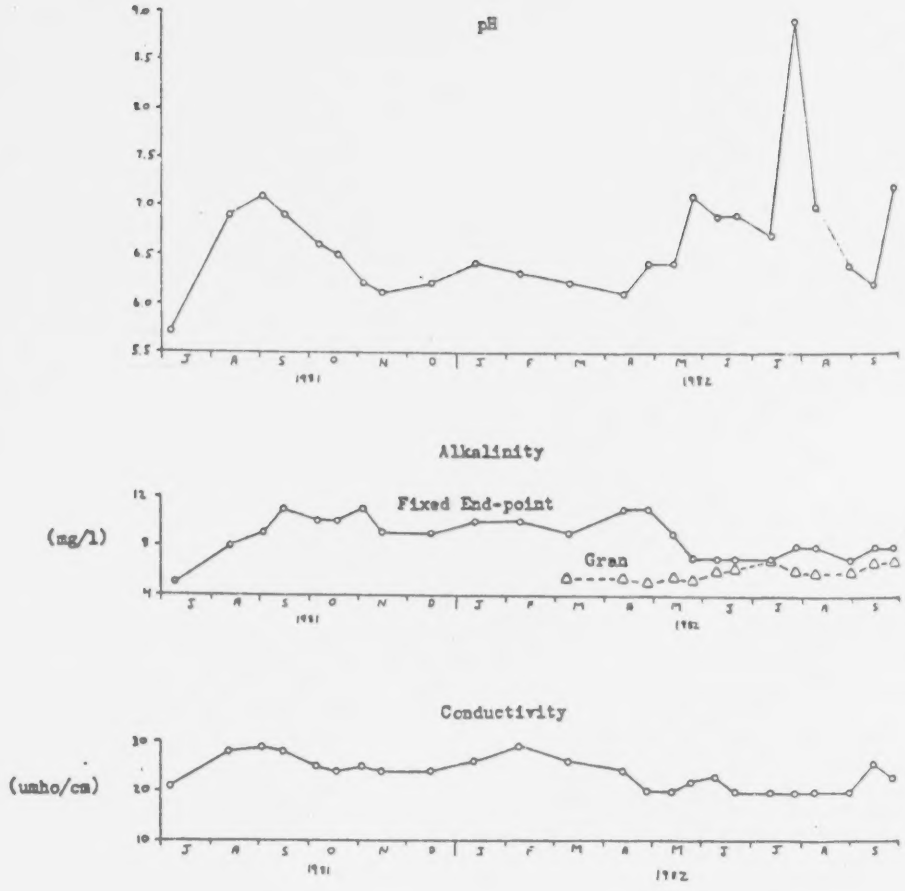


Figure 9-25. The pH, alkalinity, and conductivity trends for Round Lake.

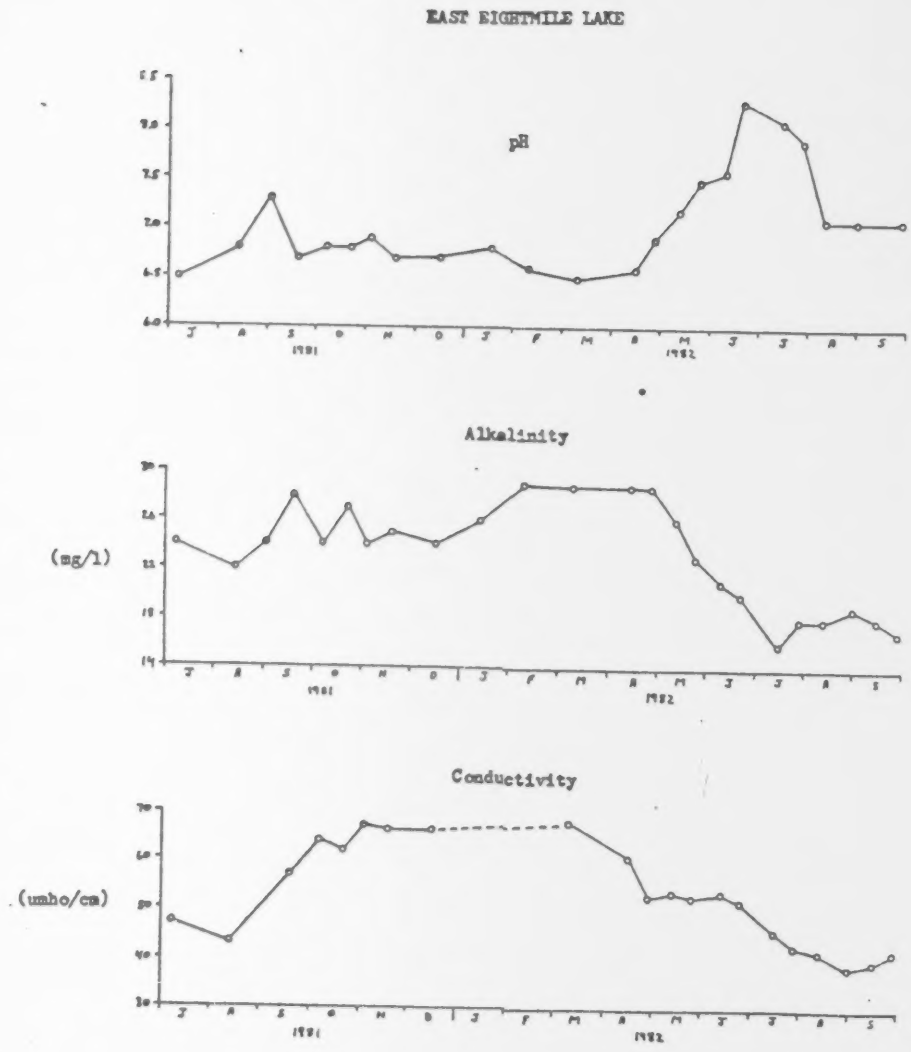


Figure 9-26. The pH, alkalinity, and conductivity trends for East Eightmile Lake.

maximum of 8.3 at the end of June, then declined to 7.1 by early August. Alkalinity in East Eight Mile Lake was higher during the ice covered period than during the summer and fall of 1981, but after ice melt alkalinity decreased from 29 mg/l to about 16-18 mg/l, which was maintained from July through September.

Conductivity followed the same seasonal trends as alkalinity in that it was high in the fall of 1981 and the following winter but decreased during the summer of 1982.

In Round Lake total phosphorus concentrations generally decreased from August 1981 until the onset of ice cover (Figure 9-27). With ice melt total phosphorus values generally increased to a high of 0.024 mg/l. This same trend occurred in East Eight Mile Lake although concentrations were lower, reaching a maximum value of 0.019 mg/l. Total nitrogen concentrations were generally highest in the fall of 1981 and the following winter but showed an increasing trend throughout the summer of 1982.

Seasonal trends in water clarity and chlorophyll a are shown in Figure 9-28. Chlorophyll values given for Round Lake are the weighted mean for the upper 10 meters, while the values for East Eight Mile Lake are the weighted mean for the whole water column. The maximum chlorophyll peak in 1981 in Round Lake was in early October although a minor peak also occurred in December. In 1982 there was a spring peak in May and a larger peak in late July.

In 1982 there was no algal bloom in September and October as occurred in 1981, therefore, the maximum chlorophyll concentration in 1982 (15.5 mg/m^3) was much lower than in 1981 (28.0 mg/m^3). Minimum Secchi readings occurred during the algal blooms - 1.5 meters in early October 1981 and 3.0 meters in late July 1982. The maximum Secchi reading for Round Lake, 10.1 meters, occurred in late winter. In East Eight Mile Lake the chlorophyll concentrations were much less, with peak concentration in both 1981 (5.5 mg/m^3) and 1982 (4.5 mg/m^3) occurring in early September. Water clarity was poorest when chlorophyll concentrations were highest (4.6 meters in 1981 and 4.0 meters in 1982).

Phytoplankton composition and biovolume illustrations for Round Lake (Figures 9-29 and 9-30) show that in 1981 total biovolume has less than 1 mm^3/l except during the algal bloom in September and early October which consisted entirely of the blue-green alga Anabaena affinis. After the 1981 algal bloom the phytoplankton assemblage was dominated by cryptomonads. In 1982 there was a spring phytoplankton pulse that was dominated by dinoflagellates and to a lesser extent chrysophytes. The only Dinophyceae present was Peridinium cinctum; the dominant chrysophyte was Dinobryon cylindricum. Unlike the summer bloom in 1981, the 1982 bloom was not completely dominated by blue-green species. Instead, dinoflagellates and green algae were subdominants. While the dominant blue-green algae was still A. affinis, the dominant dinoflagellate was Ceratium hirundinella and the dominant green algae were Ulothrix subtilissima and U. tenerrima.

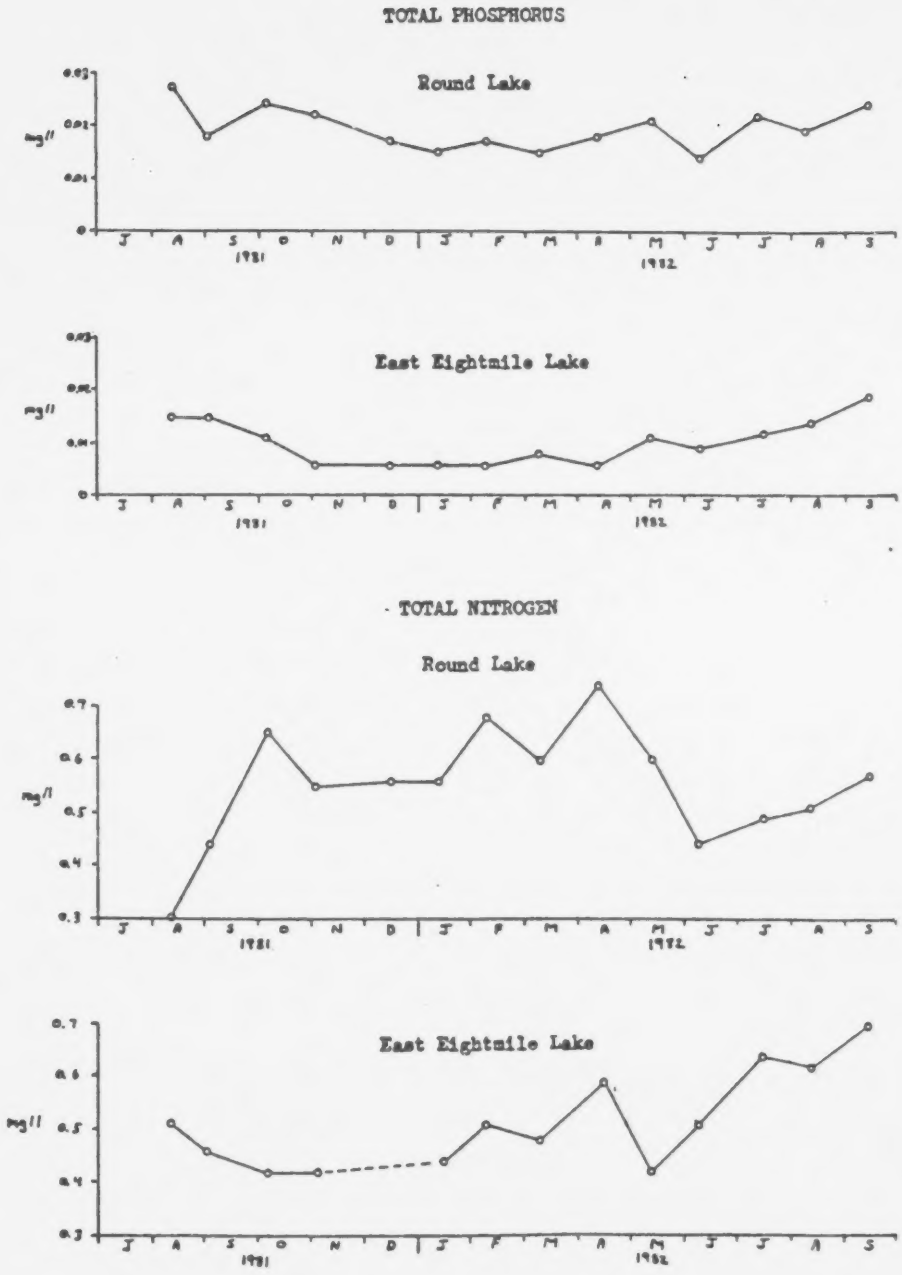
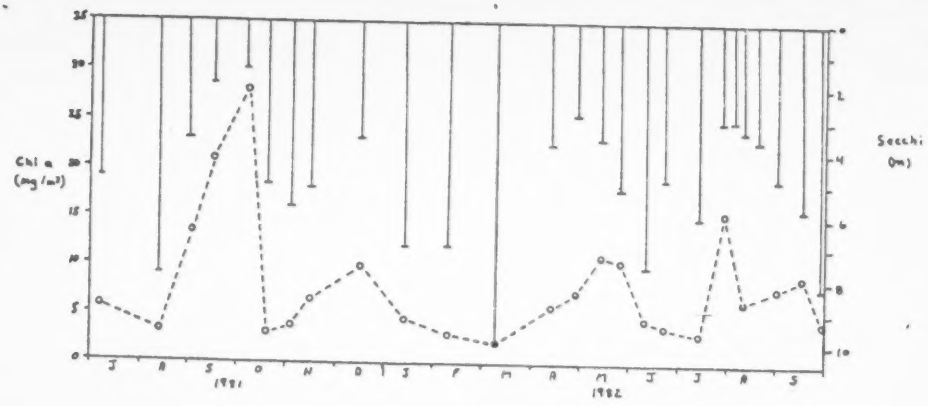


Figure 9-27. **Seasonal trends in phosphorus and nitrogen in the study lakes.**

ROUND LAKE



EAST EIGHTMILE LAKE

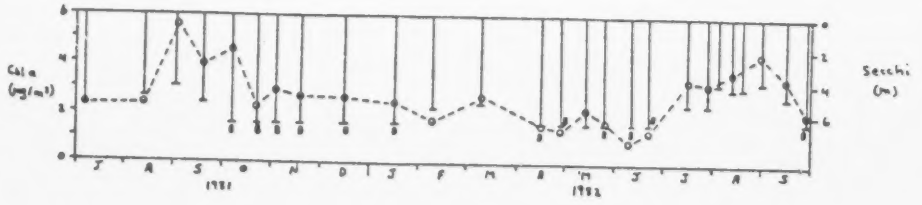


Figure 9-28. Seasonal trends of Secchi depth and chlorophyll a for the study lakes.

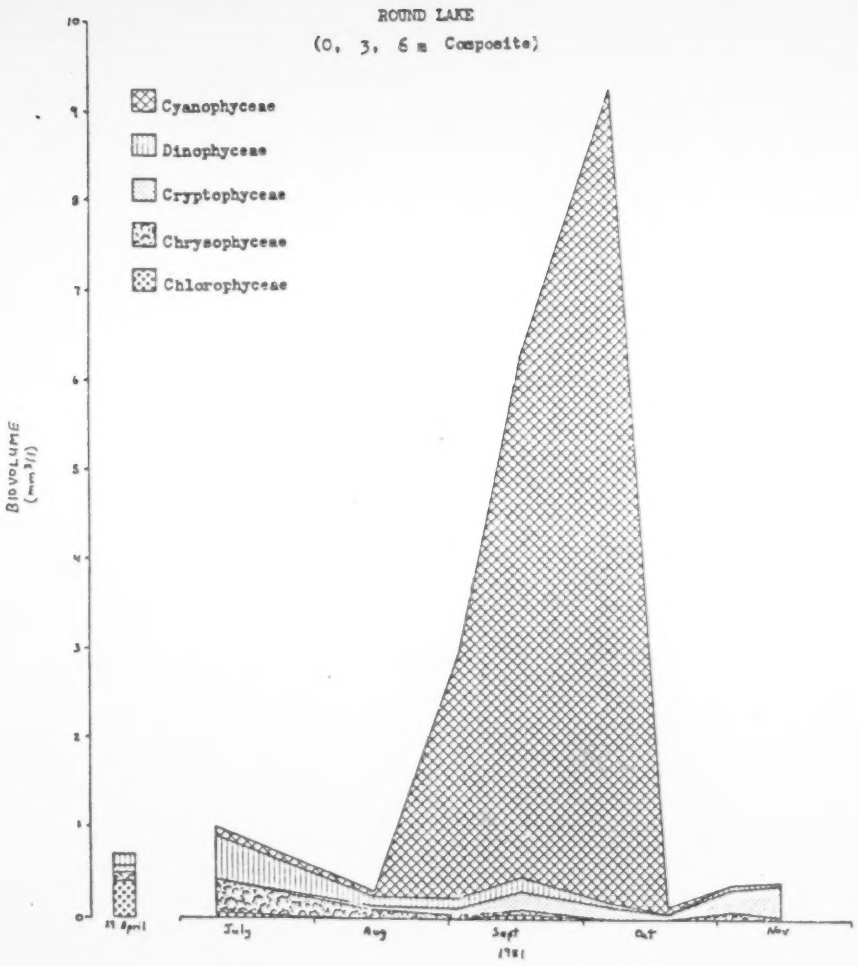


Figure 9-29. Round Lake phytoplankton composition 1981.

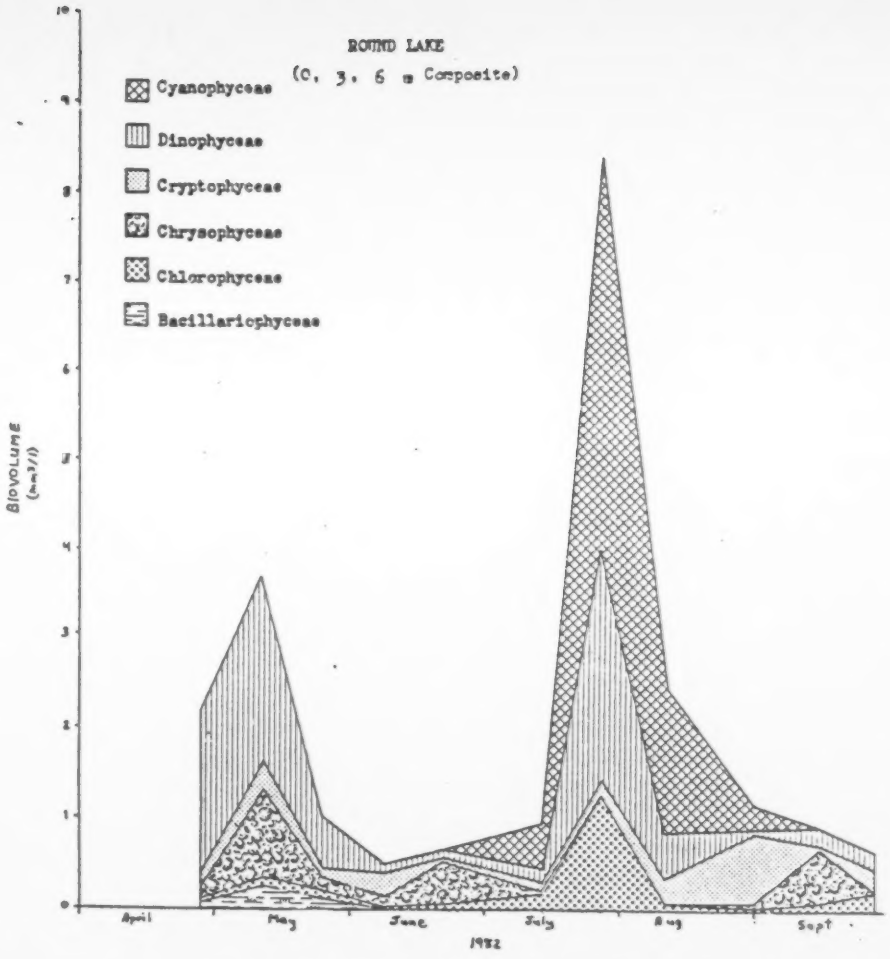


Figure 9-30. Round Lake phytoplankton composition in 1982.

The phytoplankton population in general, was much lower in East Eight Mile Lake than Round Lake. In 1981 the biovolume was usually less than 0.2 mm^3/l (Figure 9-31). However, there was an increase in the phytoplankton in September composed of blue-green algae (e.g. Microcystis flos-aquae) and chrysophytes (e.g. Uroglenopsis americana) in the early part of the month and the diatom Cyclotella comensis in mid-September.

In 1982 the dominant alga of the spring population was the dinoflagellate P. inctum. The summer algal increase was much greater and earlier in 1982 (Figure 9-32). In mid-July there was an increase in C. hirundella). This was followed in August by an increase in algal biovolume to a maximum of 3 mm^3/l. The algal assemblage was dominated by the blue-green alga M. flos-aquae and to a lesser extent, M. aeruginosa.

Seasonal composition and trend data for the major zooplankton groups in Round Lake (Figure 9-33) reveal that during most of the year the Rotifera constitute the most abundant groups, but because of their small size they are less important in the total zooplankton biomass. The Rotifera exhibited two population peaks, one in December that was predominately the Conochilus complex (including both Conochila unicornis and Conochiloides dossuarius) and another from late June through mid-September dominated primarily by Collotheca mutabilis and secondarily by Kellicottia longispina.

The Cladocera were present in low concentrations during the summer and fall of 1981 but in December there was an increase in Bosmina longirostris. This high population was maintained through most of the winter, declining in April, 1982. B. longirostris exhibited another population increase in late July and early August. The Cladoceran peak also included Daphnia galeata mendotae and Diaphanosoma leuchtenbergianum.

The dominant calanoid copepod found in Round Lake was Leptodiaptomus minutus. The population peaks were during the ice cover period and during the summer, but the summer peak was later than the Cladoceran peak, occurring in mid-September. The largest part of the population was in the naupliar stage of the life cycle.

The cyclopoid copepods exhibited a complex seasonal trend. At least four population peaks occurred throughout the sampling period. Although two species were found in Round Lake, Cyclops bicuspidatus thomasii and Tropocyclops prasinus, the dominant one was C. bicuspidatus thomasi.

Although the Rotifera were the most abundant zooplankton group in East Eight Mile Lake (Figure 9-34) they were less important than in Round Lake. The population level was fairly stable throughout the year with the exception of the late ice cover period. Cladocera exhibited peaks in August 1981, and June, July, and September 1982. The peak in August 1981 also included B. longirostris and Ceriodaphnia lacustris. The peak in June and July 1982 was mostly D. galeata mendotae, which the peak in September was B. longirostris. The calanoid copepods (Skistodiaptomus oregonensis) were generally more abundant than the cyclopoids. As in Round Lake the calanoids and Cladocera peaked at similar times. This may be because both groups are herbivores with similar nutritional requirements.

EAST EIGHTMILE LAKE
(0, 3, 6 m Composite)

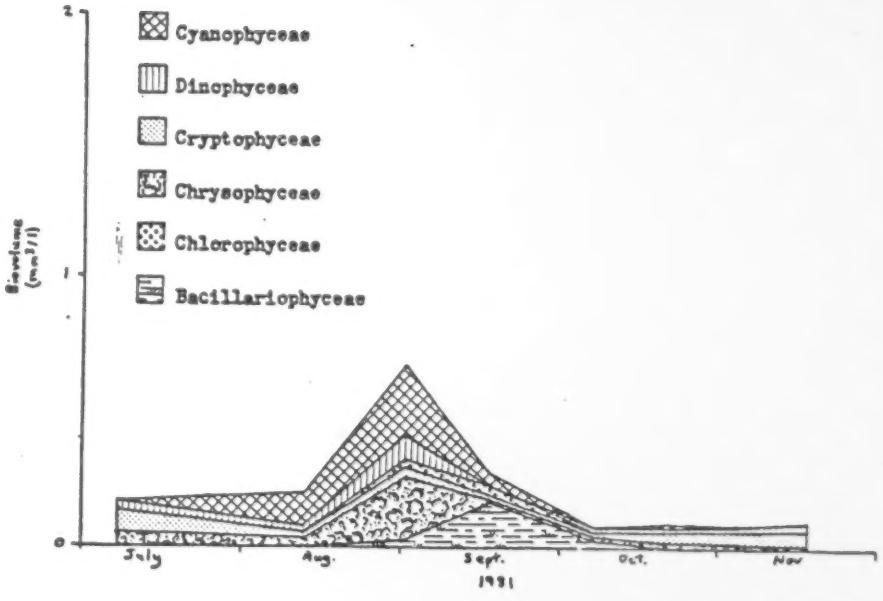


Figure 9-31. East Eightmile Lake phytoplankton composition in 1981.

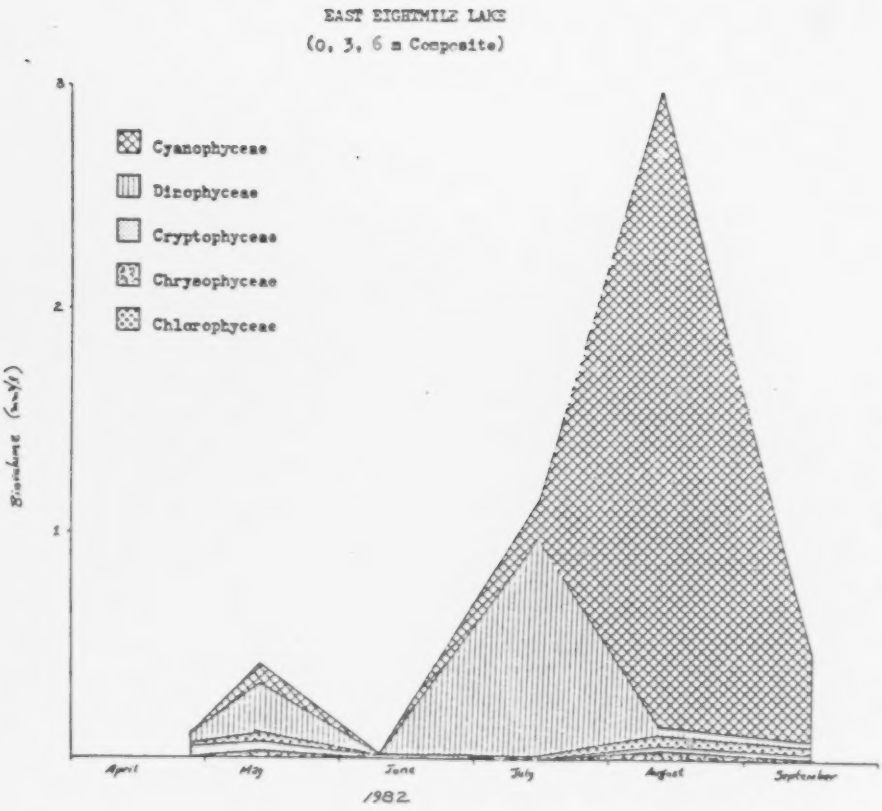


Figure 9-32. East Eightmile Lake phytoplankton composition 1982

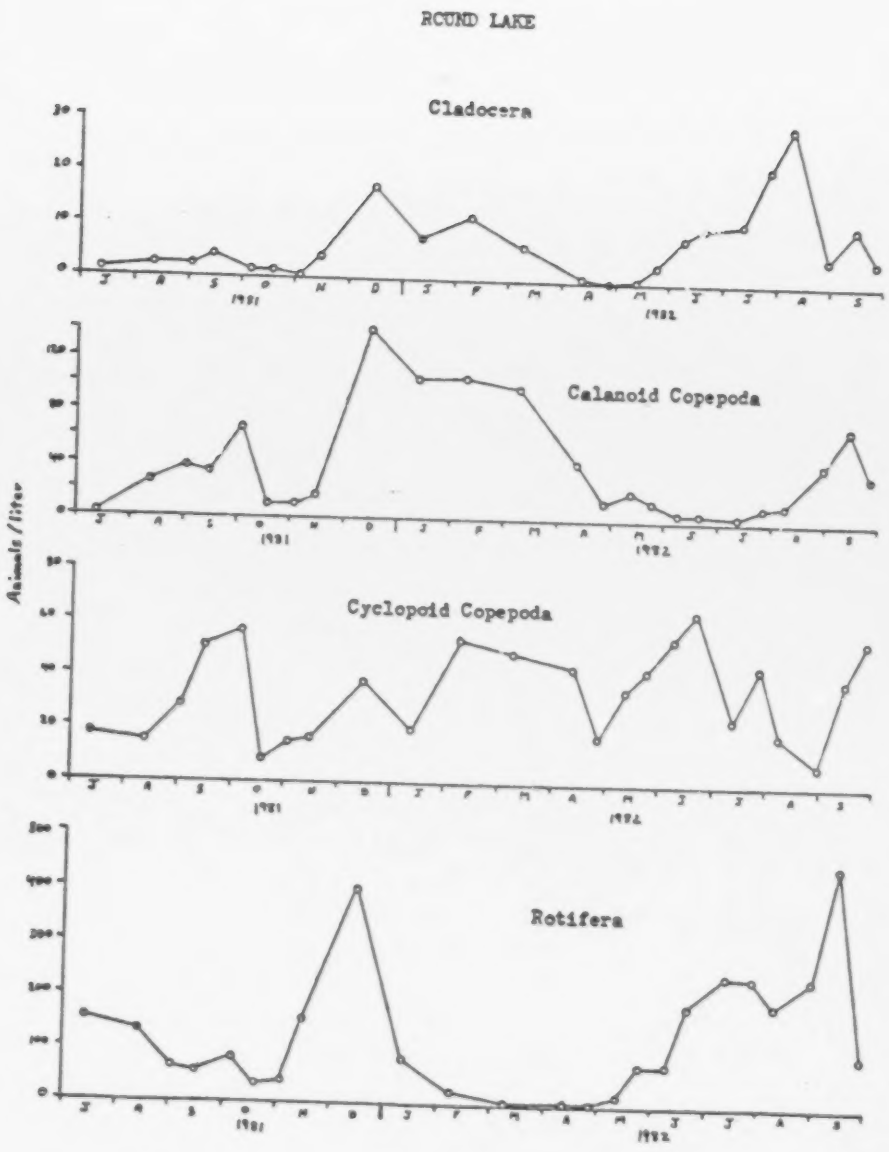


Figure 9-33. Seasonal trends in zooplankton composition in Round Lake.

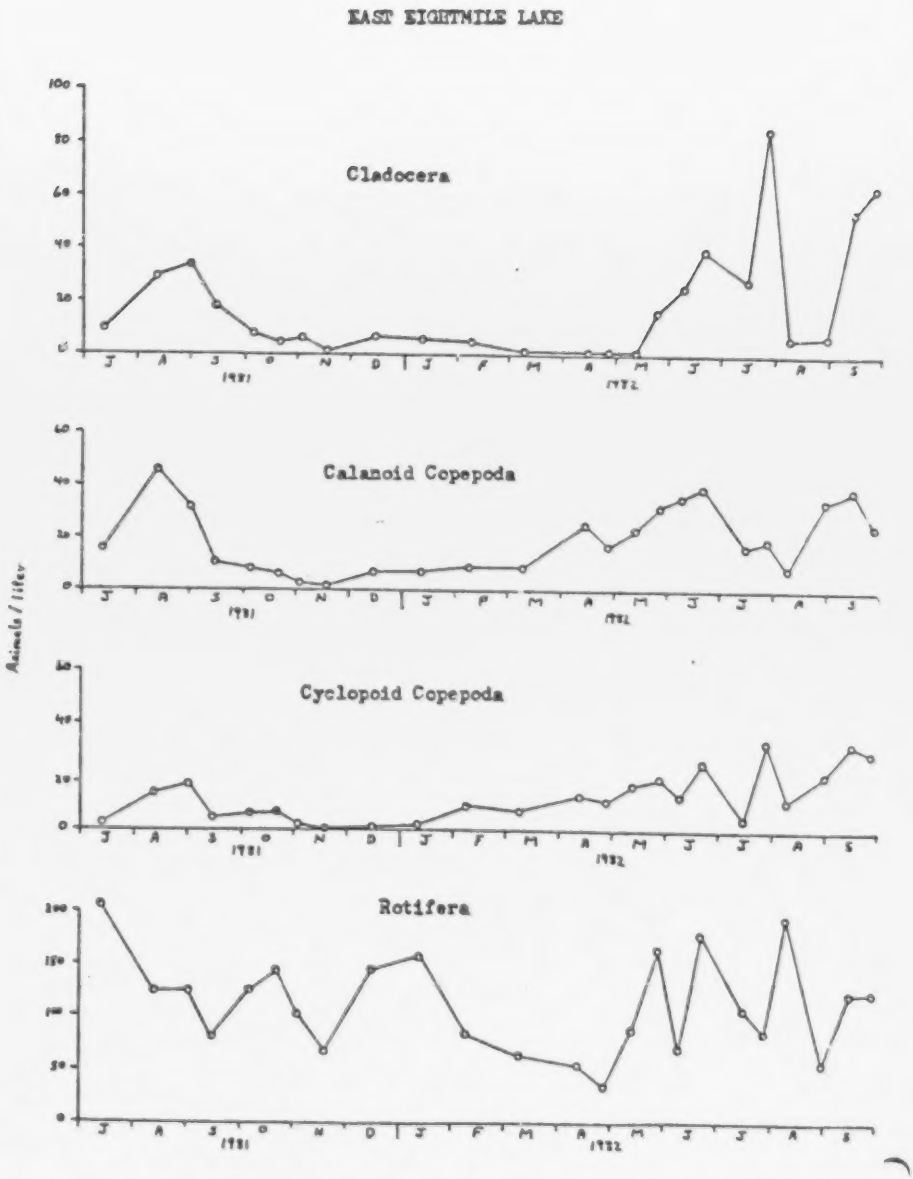


Figure 9-34. Seasonal trends in zooplankton composition in East Eightmile Lake. -110-

The macrophyte community in Round Lake is relatively simple and includes only submerged species. The community is principally segregated by substrate and to a lesser extent by depth. From 0 to about 3 meters, a sparse community of principally Eleocharis acicularis was found, and the substrate was sand. From 3 to 6 meters the substrate was still sandy but in this zone there were scattered patches of Isoetes sp. At about 6 meters the substrate became muck and the community was exclusively the alga Nitella cf. opaca. N. opaca was found occasionally at shallower depths but only where there were patches of muck. Nitella was very abundant to a depth of 10.5 meters but began to thin thereafter and was not found beyond a depth of 12 meters.

The macrophyte community in East Eight Mile Lake was more extensive, covering large areas of the lake bottom. The sparse nearshore community was made up of mostly Najas flexilus and Juncus pelocarpus and E. acicularis. At about 2.5 meters scattered patches of Potamogeton amplifolius began and extended out to about 4.5 meters. At about 4.0 meters N. flexilus was replaced by the alga Chara sp. which formed a luxuriant growth covering the deeper portion of the lake.

Benthic invertebrate samples were collected from both lakes but only the February 1982 samples have been analyzed. In Round Lake, at all stations except RLIII the population was dominated by the predacious dipteran Chaoborus punctipennis. At Station RLIII Chaoborus numbers were very low and instead the population was dominated by oligochaeta and Chironomidae. In East Eight Mile Lake the dominant group was Chironomidae; a great number of genera were present representing a very diverse population and Chironomus was one of the most abundant.

Because the mean depth of Round Lake (8.4 meters) is much greater than East Eight Mile Lake (2.9 meters), Round Lake is dimictic while East Eight Mile Lake is polymictic. This difference influences the biological and chemical limnology of the two lakes. Water temperature and dissolved oxygen differences influence the type of benthic invertebrates found in each lake; Round Lake was dominated by Chaoborus punctipennis which is a predator upon the pelagic zooplankton, whereas in East Eight Mile Lake the dominant benthic invertebrate was Chironomidae, which are generally herbivorous and omnivorous, feeding on and within the sediment. The differences in the zooplankton populations of the two lakes are also partially due to differences in their water temperature regimes. In Round Lake there were species which are found only in the cooler waters of the metalimnion and hypolimnion. In contrast, zooplankton species which usually only inhabit the littoral zone of lakes were found in the pelagic zone in Each Eight Mile Lake.

A most important difference between these two lakes is their buffering capacity. Even though Round Lake has considerably less buffering capacity than East Eight Mile, indications are that the lake has not been affected by acid precipitation. Preliminary work on a sediment core from Round Lake indicates that the dominant diatom historically was Tabellaria flocculosa. This diatom is also dominant at the present time. Both Merilainen (1967) and Tolonen (1967) consider this diatom to be

acidophilous, meaning it occurs at about pH 7 but has the widest distribution below pH 7 but above pH 5.5. The rest of the present day phytoplankton assemblage also indicates that neither lake is being adversely impacted. Kwiatkowski and Roff (1976) noted that as lakes became acidic the number of species declined, especially the Cryptophyceae. The total number of species found in 1982 was high in both study lakes, 62 in Round Lake and 52 in East Eight Mile Lake. In addition, 7 species of Cryptophyceae were found in Round Lake.

There is no evidence of any pH effect on zooplankton populations in the study lake. It was noted in Ontario that as the pH decreased the number of zooplankton declined (Sprules 1975, Roff and Kwiatkowski 1977). Both East Eight Mile and Round Lakes had 15-16 crustacean species present with 5-6 that were dominant. Round Lake is the more sensitive of the two lakes, yet it contained populations of Diaphanosoma leuchtenbergianum, Daphnia galeata mendotae and Tropocyclops prasinus, all of which are sensitive to acidic conditions (Sprules 1975). Raddum et al. (1980) found that the rotifers Keratella (exc. K. serrulata) and Conochilus were also sensitive to acidic conditions, but large populations of both of these genera are found in Round and East Eight Mile Lakes.

The macrophyte community apparently has changed in Round Lake in recent years. Fassett (1934) noted a large population of Littorella americana in that lake but it was not found in 1982 for some unknown reason. It is possible the disappearance of this species could be due to change in substrate or water levels.

The great fluctuation in pH observed in Round Lake (5.7 - 8.9) appeared to be regulated by internal rather than external factors. The highest pH's in both years occurred just prior to or during the chlorophyll peak and probably were the result of algal production removing carbonic acid from the system. Conversely, the lowest pH values occurred during low phytoplankton biovolumes. In East Eight Mile Lake the pH values steadily increased from ice out in April 1982 to a peak and plateau in June and July, which may be due at least in part to macrophyte growth. In August the macrophytes began to decline and pH also declined.

Because of the relatively low buffering capacity of its waters, Round Lake might appear to be particularly susceptible to acid precipitation. However, in addition to its apparently significant watershed buffer, the lake may also have some internal buffering systems that could help ameliorate the effects of acid precipitation. Kelly et al. (1982) found that lakes with an anaerobic hypolimnion produced alkalinity, principally as a result of microbial reduction of sulfate (as sulfate is reduced HS combines with H+ to increase alkalinity). At this time it has not been determined how important this process is in Round Lake, but it could be a significant factor.

Eight different species of fish have been found in East Eight Mile Lake, and five species are present in Round Lake, discounting stocked species (Table 9-10). East Eight Mile Lake has a population structure of northern pike, largemouth bass, and panfish that is typical of many small, shallow

lakes in northern Wisconsin. Panfish are most abundant in East Eight Mile Lake, but there are also good numbers of northern pike and largemouth bass present. The Round Lake fish population is made up almost entirely of panfish and minnows at the present time; there are very few game fish in the lake, and most of those that are there have been stocked. According to local residents, Round Lake at one time had a thriving smallmouth bass population, and many large fish were reportedly caught. Now, there are very few bass in the lake, and the population appears to be on the verge of extinction.

Table 9-10. Fish species found in Round and E. Eightmile Lakes

Lake	Species	Apparent Population Status
Round*	Smallmouth bass	Very low
	Bluntnose minnow	Very abundant
	Rock bass	Very abundant
	Yellow perch	Very abundant
	Bluegill	Very abundant
E. Eightmile	Northern pike	Abundant
	Largemouth bass	Abundant
	White sucker	Low
	Golden shiner	Low
	Yellow perch	Very abundant
	Pumpkinseed	Low
	Bluegill	Very abundant
	Rock bass	Abundant

*Also has stocked trout and walleyes

10. Aquatic Resources Susceptibility Project

Introduction

The purpose of this project is to determine the susceptibility of northern Wisconsin lakes to damage from acid deposition. The approach consists of assessing the current susceptibility of area lakes, determining lake changes based on comparisons with historical data, and predicting impacts to the aquatic resources.

Lake susceptibility is being evaluated by sampling a representative number of area lakes. Over 400 lakes in the region have been sampled for water chemistry to date, many of these with multiple samplings. Understanding the biota in these lakes has also been enhanced through extensive sampling of fish, zooplankton, and other biological components. Over 100 lakes in the region have recently been sampled for biota.

Our approach in determining water quality changes in lakes has progressed in a manner similar to that used for quantifying susceptibility of area lakes. This method has consisted of determining large-scale patterns and trends by observing general responses in large populations and focussing in on smaller sample sizes. This approach has the advantage of providing an early understanding of the range of responses to be expected and allowing us to make an educated selection of lakes for more intensive study. Consequently, our "first-cut" approximation of water quality changes is based on a sample size of 175 lakes, while our more intensive subsequent work is restricted to only 60 lakes.

The foundation for predicting impacts from acid deposition in the study area is currently being set in place. The study assessing factors influencing susceptibility will provide some of the necessary information on parameterization of the 'model'. Much of the remaining information required to develop a predictive tool for the region is expected from the intensive data gathering on Clara and Vandercook Lakes and their respective watersheds.

The necessary high quality atmospheric deposition data supporting all of the above activities continue to be collected at the NADP stations and, now, at the Clara and Vandercock study sites.

The emphasis for the project is shifting from one of primarily data gathering to one of data consolidation and analysis. Although field sampling is expected to continue, the scope and intensity of all monitoring efforts is decreasing. The data gathered on our intensively monitored lakes, Clara and Vandercook, indicate that the frequency of sampling for most seasons can be reduced and still obtain the necessary data for completing the hydrologic and chemical budgets. Questions raised by the Birge and Juday data comparison will necessitate a short-term (about two weeks) resampling of some study lakes, but this program will represent only 15% of the effort required to complete the original 1980 survey.

The North Central District (NCD) began investigating the susceptibility of area lakes to acid deposition in the summer 1979 when 275 lakes were sampled. This investigation was carried out in partnership with the Environmental Protection Agency (EPA) through their Environmental Research Laboratory in Duluth (ERLD). As part of this joint effort, numerous studies have developed to address questions raised early in the program. The fundamental questions being addressed are:

1) How susceptible are the northern Wisconsin lakes to cultural acidification?

2) What are the factors determining lake susceptibility?

3) What is the composition of the precipitation?

4) Have the lakes become more acidic as a consequence of acid deposition?

5) What would be the biological consequences of acidification of these lakes?

Project supervisor for the susceptibility studies being conducted by the Department of Natural Resources in its North Central District, is Robert Martini. This work is well coordinated with the six projects previously described. Project organization, responsibilities, and relationship to the Joint Acid Deposition Technical Review Committee are shown in Figure 10-1.

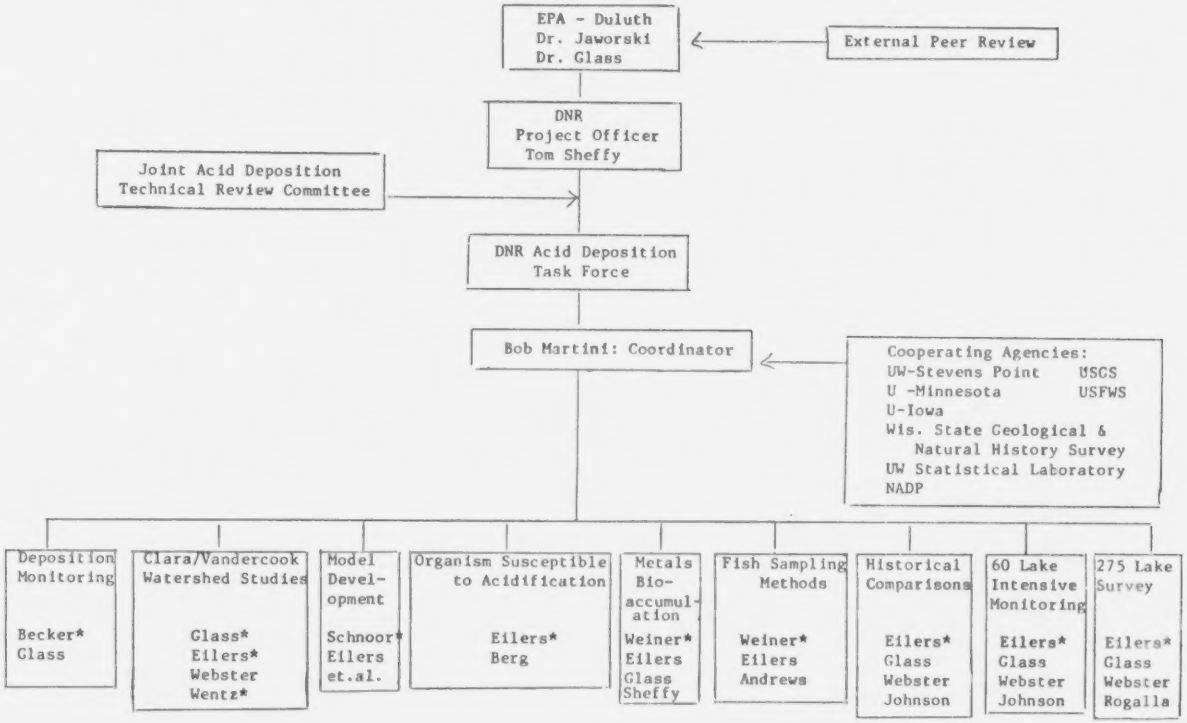
Each of these major areas of investigation have been addressed to date although work continues in most areas. The number of institutions involved in this work has expanded to include (in addition to the WDNR and EPA) the following: U.S. Geological Survey (USGS), University of Minnesota (UM), University of Wisconsin (UW), University of Iowa (UI) and the U.S. Fish and Wildlife Service (USFWS). The primary interactions of NCD and these organizations are shown in Table 10-1. The individual projects are summarized in the following section. Based on information from the literature, a general comparison has been made between susceptibility of Wisconsin lakes and lakes in the Adirondeck Mountains of New York State (Table 10-2).

Project Summaries

275 Lake General Survey - The primary purpose of this project was to determine the general water quality of a representative group of area lakes. Over 275 lakes (approximately 10% of area lakes) were sampled once in summer 1979 to provide an adequate sample size. Samples were split between DNR and EPA (ERLD).

DNR sampling was financed by an areawide water quality planning grant for the Upper Wisconsin River Basin.

Figure 10-1. Table of Organization and Reporting Structure for Aquatic Resources Susceptibility Project.



* Denotes Principal Investigators

Table 10-1 Cooperation of NCD and Other Institutions Related to Acid Rain Research in North Central Wisconsin.

	Project Description	USEPA	USGS	USFWS	UW	UM	UI
1	General Lake Survey (1979 275-lake sampling)	*Split Sample Analysis *Cations				*Review (D)	
2	Intensive Lake Survey (1980 60-lake monitoring)	*Split Sample Analysis *Cations/Anions				*Anion Analyses (M)	
3	Historical Comparisons (Comparisons with Birge & Juday Lake data)	*Services thru Projects 1 and 2		*Some support for keypunching historical data (M)	*Statistical support (Statistical laboratory	*Anion Analyses (M)	
4	Deposition Monitoring (NADP, bulk deposition and snow surveys)	*Provided bulk samplers *NADP assistance	*Bulk sampling in winter 1981-1982				
5	Clara/Vandercook (hydrologic, chemical mass balance of two soft water lakes)	*Water chemistry analysis	*Water budget	*Fish sampling Clara	*Fish Sampling Vandercook (M) *Zooplankton (SP)		*Trickle-down model testing on both lakes
6	Model Development (chemical response model of acidification)	*Some financial support to UI	*Will provide hydrology for Clara/Vandercook			*Model first tested in Minnesota (D)	*Lead organization
7	General Organism Susceptibility (literature review)	*Literature support		*Review	*Review	*Review (M)	
8	Metals Bioaccumulation (Mercury uptake related to water chemistry)	*Providing water chemistry		*Lead Agency w/DNR			
9	Fish Sampling Methods (analysis of sampling fish in ultra-soft water lakes)	*94-lake seining survey		*12-lake multi-gear sampling *lead agency			

M = Madison
SP = Stevens Point

D = Duluth
M = Minneapolis

Table 10-2. SUSCEPTIBILITY OF WISCONSIN LAKES COMPARED TO LAKES IN THE ADIRONDACKS

Characteristic Being Compared	Adirondacks	Wisconsin
1. Location	First uplands downwind of major sources	Western edge of "acid rain belt"
2. Watershed relief	Steep slope	Gradual slope
3. Elevation	2000 - 4000 ft.	1000 - 1500 ft.
4. Total rainfall	45 in/yr	30 in/yr
5. pH of precipitation	4.1	4.6
6. Sulfate loading	40 - 60 kg/hectare/yr	20 kg/hectare/yr
7. Bedrock	Granite (Frequent outcropping)	Granite/sandstone
8. Drift (soil) thickness over bedrock	Thin (A few meters)	Thick (10 - 100 meters)
9. Drift characteristics	Sandy	Sandy/sparse calcareous deposits

Sampling is completed. A presentation of these results was made to the 1982 national meeting of the American Chemical Society. A manuscript was accepted for publication by the Canadian Journal of Fisheries and Aquatic Sciences. Additional work is proposed for FY 84 (contingent on additional funding) on relating metal concentrations to watershed and lake chemistry factors.

One third of the study lakes were considered to be highly susceptible to acidification. The primary factor controlling susceptibility in this area is hydrology, with seepage lakes being the most susceptible.

This is a qualitative assessment of lake susceptibility. The next step is to quantify the relationship between hydrology and susceptibility.

60 Lake Intensive Monitoring - The 275 lake survey in 1979 confirmed that area lakes were highly susceptible. Discovery by our staff of the raw historical lake data collected by the Wisconsin Geological and Natural History Survey and UW (Birge & Juday) made it possible to compare present water quality with historical values. We worked with University of Wisconsin-Madison (Statistical Laboratory) in designing an effective restudy of selected softwater lakes which we executed in summer 1980. This monitoring was also designed to address other questions related to lake acidification not necessarily linked to the historical comparison.

Some of the early results were presented at the 1982 national meeting of the American Chemical Society. All cations have been analyzed; anions are currently being analyzed. We have incorporated a portion of these results into a joint data set with those lakes in the 275 lake set common to the Birge and Juday set (see following project for details). The work not related to the historical comparison awaits completion of the anion analyses. A manuscript describing the second aspect of this project is planned for June 1983.

See the following project for historical comparison results. The remainder of the work demonstrates that the spatial variability of most parameters in the surface water is within the error of measurement. Road salting is having an important impact on lakes receiving highway runoff. Current empirical models predicting lake stratification do not satisfactorily predict depth of thermocline.

Historical Comparisons - Birge and Juday organized a sampling of 608 northern Wisconsin lakes from 1925-1941. This data set provides an opportunity to compare present lake conditions with those of fifty years ago. The recent data sets being compared with the historical data set consist of a merged data set composed of lakes sampled in 1979 and 1980.

The first stage of the historical comparison is almost complete. A presentation was made at the 1982 national meeting of the American Chemical Society. A manuscript is in preparation and is scheduled to be submitted to a journal in March 1983 (Eilers, Glass, Pollack and Loucks).

Most of the 161 lakes in this comparison have increased in pH, alkalinity and conductivity. Few of the changes in pH are significant because of the large variability associated with this parameter. The largest increases in conductivity are caused by road salting. The increases in alkalinity are in many cases associated with corresponding increases in lakeshore development.

Deposition Monitoring - Interest in the quality of regional precipitation was heightened in 1979 with the results of our general lake survey. In January 1980, the NCD established an NADP deposition monitoring station at Trout Lake, Vilas County.

The results were presented at the 1982 national meeting of the American Chemical Society. The early NADP monitoring has since been supplemented by bulk deposition sampling at a variety of sites carried out by EPA, NCD and USGS. In addition, EPA and NCD have carried out spring snowpack sampling in 1980-82. Becker, et al (1982) presented a paper at the American Chemical Society National Symposium on Acid Rain in Las Vegas, Nevada.

The annual volume-weighted pH of precipitation averages from 4.6-4.7. The snowpack tends to be slightly less acidic.

Clara and Vandercook Watershed Studies - The in-lake sampling programs carried out previously are insufficient for developing and testing mechanistic models of lake acidification. Consequently, DNR-NCD developed a joint program with the USGS and EPA to gather the necessary information to provide hydrologic and chemical mass balances for two lake watersheds, Clara and Vandercook lakes. Data collection began in Fall 1980.

The early results were presented at the 1982 Wisconsin Chapter of the American Water Resources Association meeting. The groundwater model for Vandercook Lake is operational and closure on the water balance is expected in March 1983. The hydrology for Clara Lake will be completed in June 1983. The chemical balances for the two systems will follow shortly. The University of Iowa has selected Clara and Vandercook as the test sites for the trickle-down model. Zooplankton and macrophyte analysis of the two lakes will be completed by April 1983. Lakes Clara and Vandercook will probably be selected as study sites for four graduate students from two campuses of the University of Wisconsin.

Lakes Clara and Vandercook are primarily groundwater discharge lakes with relatively long residence times. Although both lakes have very similar chemistry (pH 6.1, alkalinity 20-40 ueq/l), some fish in Lake Clara show elevated (e.g. 1 ug/gm) levels of mercury.

Model Development - The development of the "Trickle-Down" model by Dr. J. Schnoor (Univ. of Iowa) is intended to fill the large gap between the emperical models (e.g., Henriksen) and the data-intensive models such as the one developed by Tetra Tech in the ILWAS project. We began to work with Dr. Schnoor in 1981 who also was receiving support from EPA at that time.

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The model was first developed and tested in the Boundary Water Canoe Area of Minnesota. The baseline work for testing the model on Clara and Vandercook has been completed and it can be evaluated with the completion of the water budgets by USGS this spring. Several presentations have been made and a manuscript describing the model has been submitted for publication (Schnoor, Eilers and Glass).

The model shows that within a hydrologic type, weathering rate is a key factor in determining lake susceptibility. If laboratory measurements of weathering rates are applicable to field conditions, then a two-fold increase in acidity of precipitation would cause decreases in pH of more than one unit for approximately 25% of precipitation-dominated lakes which we observed.

Organism Susceptibility to Acidification - Generalizations about aquatic community response to acidification involve assumptions about intra-species variability. DNR-NCD conducted a literature review of minimum observed pH values for over 1900 aquatic organisms.

The computer file has been compiled and an internal document describing the results has been prepared (Eilers and Berg). The manuscript will be modified to focus more on organisms from Wisconsin and may be submitted for publication. A presentation of this material was made at the American Chemical Society National Acid Rain Symposium in Las Vegas, March 1982.

There are wide variations in individual species response to low pH which make it difficult to predict what might be the biological impacts to Wisconsin lakes based on observations from Ontario, Scandanavia and New York. Nevertheless, it is apparent certain groups of organisms, particularly the molluscs, will be eliminated long before a measurable effect can be observed on the fish community.

Metals Bioaccumulation - Dr. Jim Wiener (USFWS-LaCrosse) intensively sampled the fish populations of 12 lakes in North Central Wisconsin during the summer of 1980. The results of the mercury concentrations in fish flesh were made available to WDNR in spring 1982. This data revealed elevated concentrations of mercury, in a subset of the study lakes. The metal concentrations showed a strong negative correlation with pH. The WDNR followed up the USFWS study with a sampling program of 19 lakes that spanned a broad range in pH.

Sixty samples from 109 fish have been analyzed for mercury by the State Lab of Hygiene. The USFWS and WDNR data sets have been merged on a computer file and the manuscript has been outlined by Wiener. A draft manuscript is targeted for June 1983.

The WDNR data compare favorably with the USFWS data. Mercury accumulation was highest in walleye and least in yellow perch. Again, mercury concentrations were highest in low pH lakes.

Fish Sampling Methods - Adequately sampling fish populations has always been a difficult task. However, it is important that the fish populations

be well described when conducting acid rain related investigations. Two fish sampling programs were carried out in summer 1980. A 94-lake seining survey by EPA/WDNR and a 12-lake multiple-gear sampling by USFWS. The NCD also began investigating gear-type efficiencies by sampling six softwater lakes in 1980. This sampling program has continued into 1983.

The data sets have been placed in a computer file and data analysis is underway. A draft manuscript is anticipated in April 1983 (Wiener, Andrews, Eilers and Rago).

Adequately sampling of fish populations requires careful consideration of target fish species, fish behavior, lake morphometry and gear selectivity. No single gear type proved entirely efficient in sampling these softwater lakes.

11. Budget

Table 11-1 itemizes the individual costs of the seven research projects that make up the Wisconsin Acid Deposition Research Program. The source of funding is also shown, as well as the amount expended for each project through 12/31/82.

Wisconsin Acid Deposition Research Program

Research Component	Estimated Cost	Wisconsin Utility Association Commitment	Wisconsin DNR Commitment	Other Commitments	Total Commitments To Date	Amount Expended Through 12/31/82	Balan Availa
Deposition Monitoring Project	$ 215,00	$238,790	$ 0	$ 0	$ 238,790	$ 93,530	$145,2
Deposition Data Integration and Quality Assurance Project	$ 55,000	$ 55,000	$ 19,905 *	$ 0	$ 74,905	$ 34,840	$ 40,0
Atmospheric Trajectory Project	$ 110,000	$110,000	$ 0	$ 0	$ 110,000	$ 14,129	$ 95,8
Trajectory Quality Assurance Project	$ 25,000	$ 25,000	$ 25,407 **	$ 0	$ 50,407	$ 50,407	$
Emission Sources Inventory	$ 14,500	$ 14,500	$ 9,766 **	$ 0	$ 24,266	$ 9,766	$ 14,5
Intensive Watershed Study	$ 900,000	$123,000	$154,372	$ 4,000[1] $ 44,908[2] $218,800[3] $363,649[4]	$ 908,729	$528,097	$380,6
Aquatic Resources Susceptibility Project	$ 300,000	$ 0	$ 11,703	$187,963[5]	$ 199,666	$137,057	$ 62,6
Miscellaneous Contingency Fund	$ 5,000	$ 5,000	$ 0	$ 0	$ 5,000	$ 0	$ 5,0
Program Administration	$ 37,500	$ 37,500	$ 52,000 **	$ 0	$ 89,500	$ 27,116	$ 62,3
	$1,662,000	$608,790	$ 273,153	$819,320	$1,701,263	$894,942	$806,321

*Data processing support and staff.

**Staff time.

[1]Wisconsin Geological and National History Survey
[2]University of Wisconsin, Madison -- Soils Department
[3]United States Geological Survey
[4]Electric Power Research Institute
[5]U.S. Environmental Protection Agency

12. Summary

1. Acid deposition is occurring in Wisconsin. Based on 1982 precipitation data from the National Atmospheric Deposition Program sites and the Wisconsin Acid Deposition Research Program sites, the range of monthly, mass/volume weighted mean lab pH of wet deposition was:

Round Lake (Douglas County)	4.80-4.81
Spooner (Washburn County)	4.71-5.13
Trout Lake (Vilas County)	4.10-4.69
Lake Dubay (Portage County)	4.57-5.04
Legend Lake (Menominee County)	4.06-4.88
Lake Geneva (Walworth County)	4.31-4.70

The range of monthly sulfate (SO_4) and nitrate (NO_3) wet deposition in kilograms per hectane (Kg/Ha) was:

	SO_4	NO_3
Spooner	.05-2.03	.07-1.28
Trout Lake	.07-3.05	.12-1.91
Lake Dubay	1.57	1.20
Legend Lake	1.14-2.06	.65-1.76
Lake Geneva	1.86-4.24	1.14-2.24

These data suggest that rainfall in southeastern Wisconsin (Lake Geneva) is more acidic than rainfall in northwestern Wisconsin (Round Lake). Additionally, the loading rates of sulfate and nitrate are generally higher in the southeastern part of the state than in northwestern Wisconsin.

This is consistent with the findings of Becker, et al. 1983 (Implications of the Gradient in Acid and Base Deposition Across the Northern Lake States, Envir. Sci. and Tech., In Press) who reported that precipitation pH across the upper midwest ranges from above 5 in western Minnesota to approximately 4.3 in lower Michigan.

2. Because trajectory analyses have not been completed on a sufficient number of individual storms and other precipitation events that cause acid deposition in Wisconsin, no conclusions can presently be drawn on what proportion of the acid deposition in Wisconsin is due to in-state versus out-of-state sources.

3. Major sources of sulfur dioxide (SO_2) and nitrogen oxides (NO_x) have been identified in Wisconsin. Over 97% of Wisconsin's anthropogenic SO_2 emissions come from 43 point sources--primarily coal-fired power plants and paper mills. Over 88% of Wisconsin's point source emissions of NO_x come from 24 sources. Major sources of these pollutants outside Wisconsin need to be identified. In Wisconsin, natural sources of SO_2 are considered negligible.

4. Data collection is continuing at the Round Lake and East Eight Mile Lake study sites. To date, there is no evidence of acid rain damage in either watershed.

5. Studies in northern Wisconsin indicate that approximately 30% of the lakes in the northern one-third of the state are susceptible to acid deposition. This 30% (about 2,000 in number) generally includes the small, seepage lakes (no inlet or outlet) that receive little recharge from groundwater, or groundwater recharge with low alkalinity. These lake types have a limited ability to neutralize acid. Much of their recharge water comes from precipitation, and their chemistry may be influenced in time by this source of acidic recharge. However, many of our northern lake systems are provided with some of the minerals needed to neutralize acidity due to glacial till, groundwater discharge, and forest vegetation. The variable geology of Wisconsin puts it in the transition zone between high and low susceptibility to acid deposition.

6. A historical comparison has been made between lake water quality in 1925-1941 to lake water quality in 1979-1980. Most of the 161 lakes in this comparison have stayed the same or increased in pH, alkalinity and conductivity. However, a few of the small, low-alkalinity seepage lakes have shown a significant decline in pH and alkalinity. The largest increases in conductivity are caused by road salting. The increases in alkalinity are in many cases associated with corresponding increases in lakeshore development.

7. No damage to aquatic biota has been verified in Wisconsin lakes from acid deposition to date. Baseline surveys of lakes and aquatic life (zooplankton, macrophytes, fish) will be used for future comparison.

8. Fish have been sampled from approximately 20 lakes which span a broad range in pH. Preliminary data show elevated levels of toxic mercury in fish from low-pH lakes. This suggests that if cultural acidification were to lower the pH of surface waters in Wisconsin, toxic metals would be likely to build up in some fish.

9. Geographically, Wisconsin lies on the western fringe of the acid deposition impact region of northeastern United States. Wisconsin receives less total rainfall (30 in/yr) and less acidic rainfall (pH 4.6) than affected areas of New England that receive up to 45 inches of precipitation each year with a pH of 4.1.

10. Research findings from other geographic areas where acid deposition damage has occurred must be cautiously applied to Wisconsin because of our unique position in terms of geographic location and geologic variability. The susceptible resources in Wisconsin are quite different from those in other affected areas. The manner and rate in which Wisconsin's resources respond to acid deposition is likely to be different than the response observed elsewhere.

11. When Wisconsin lakes are compared to lakes in the Adirondack Mountains (where acid rain damage has been documented), major differences exist in terms of the susceptibility of the two geographic regions. The fact that Wisconsin has not yet observed damage from acid rain may be due to these major differences in susceptibility.

12. In view of our present understanding of the problem, Wisconsin does not face an "acid rain crisis", but future long-term damage could occur if levels of acidity remain unchanged or increase. These concerns are manifest in the comprehensive research program now underway.

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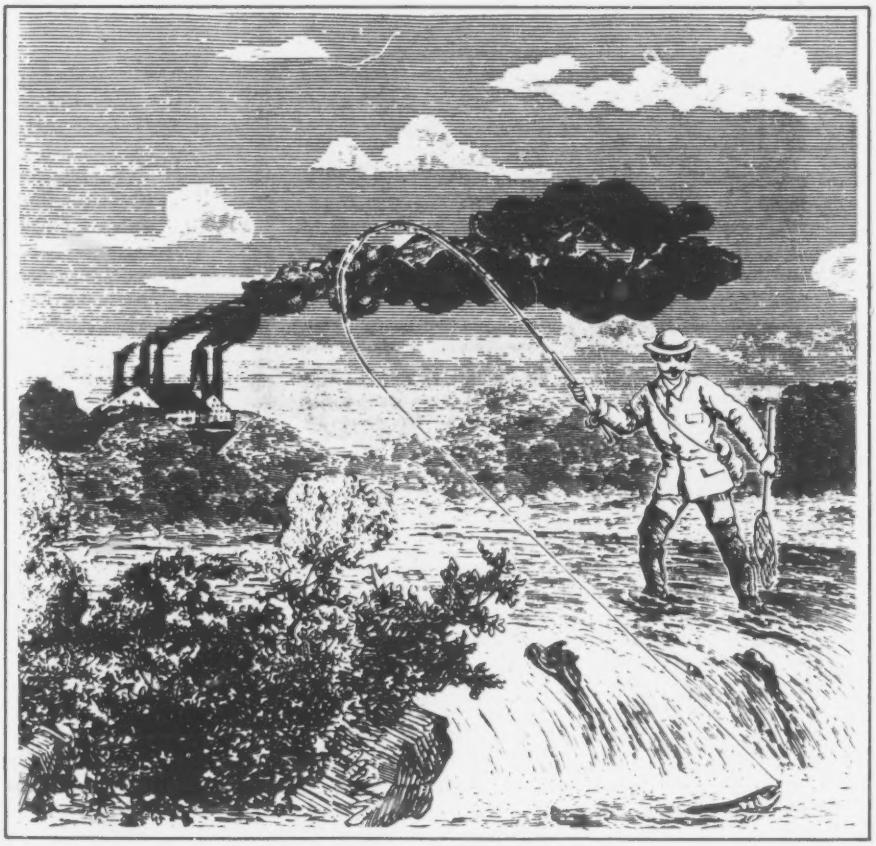
Selected strategies to reduce acidic deposition in the U.S.

Several SO_2 emission reduction strategies are examined and their attributes clarified. The effectiveness of each method is measured by the changes it produces in total sulfur deposition.



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Although the research needed to fully characterize acidic deposition and its range of effects is many years from completion, political initiatives are being taken today to formulate control programs aimed at reducing the emissions of atmospheric pollutants believed to be responsible for environmental damage. Information on what effects such emission reductions would have on acidic deposition is incomplete, and there is no certainty that decreases in emissions would significantly reduce the adverse effects of acidic deposition. Nevertheless, the political process is moving ahead rapidly at both national and international levels and may be outrunning the pace of scientific research.

This article is an attempt to shed light on this controversial policy issue by examining the attributes of several plausible acid deposition control strategies for the U.S. It discusses the costs of these strategies, the emission reductions that would be obtained, and certain secondary effects they would have on selected regions of the country. Rather than attempting to complete the cost–benefit equation—which is arguably impossible given the current state of knowledge—this study provides some measure of the potential effectiveness of these strategies by estimating consequent reductions in sulfur deposition. It describes a systematic approach to portions of an organized cost–benefit analysis of acid rain legislation, but it is not intended to provide the definitive answer to the problem. (Ecological benefits are not even addressed here.) Many of the components necessary for a complete assessment are being developed in various government and utility studies.

Despite the fact that acidic deposition and its effects are not fully understood, it is clear that certain types of strategies could be implemented to reduce precursor emissions or otherwise mitigate effects. This article emphasizes the approach that has the greatest chance of being implemented in the U.S.: continuous reduction in emissions of sulfur dioxide from electric utility power plants. While this is the most likely approach, other options are also discussed briefly: a decrease in nitrogen oxide emissions, a reduction in industrial or transportation emissions, intermittent fuel switching, and receptor mitigation (lake liming). The study examines several hypothetical sulfur dioxide control strategies, as well as other strategies that are currently being debated by congressional committees. Results are presented for the year 1995, based on the expected outcome of a 10-year program instituted in 1985. It is hoped that this analysis will contribute to the establishment of economically and environmentally sound policies for dealing with acidic deposition.

Analytical approach

To accomplish the objectives of this analysis, projections are made of the effects of various emission reduction strategies, each of which is implemented in the coal-fired electric utility power plant sector. The year 1980 is used as a base year; against this base are compared certain effects the following five alternative approaches would have in the year 1995:

- no specific acidic deposition control strategy (B95),
- a mandatory coal washing program (CWM),
- a ceiling on allowable SO_2 emissions of 2 lb $SO_2/10^6$ Btu (CP2),
- the Stafford bill, S. 3041 (STF), and
- the D'Amours bill, H.R. 4816 (DAM).

These approaches were chosen for close examination because they represent different conceptual approaches to a control program and because they cover a wide range of emission reductions. Other hypothetical approaches and congressional bills are discussed at appropriate points in the text.

The first three of these strategies—B95, CWM, and CP2—were analyzed in a joint study performed for the EPA, the Department of Energy (DOE), and Argonne National Laboratory by Teknekron Research, Inc. (*1*). In the Teknekron study, the Utility Simulation Model (USM) was used to project the effects of several control strategies for electric utilities (*2*, *3*).

The USM consists of a number of interconnected computer modules and data sets that simulate decisions involved in utility system planning and operation and the impact of these decisions on utility finances and the environment. The model is driven by a set of exogenous parameters that include electricity demand, financial market conditions, fuel characteristics and availability, the use of advanced technologies, and environmental regulations. It simulates the joint operation of the various types of generating units and various capacity classes owned by all the utilities within a state. Output from the model includes fuel use, electricity generation, capital and equipment requirements, releases of environmental pollutants, financial statistics for utility firms, and average electricity prices.

The results of some of these control strategies were published earlier (*4*), and this article draws on these results for costs and secondary impacts (*1*, *4*). State-level emission reductions for B95, CWM, and CP2 were calculated separately for use in long-range transport modeling (*5*). The USM results were obtained in 1980 and 1981 using the original version of the model employed by Teknekron Research, Inc., for EPA and DOE. The Universities Research Group on Energy (URGE), based at the University of Illinois at Urbana-Champaign, is currently designing an improved version of the USM called the Advanced Utility Simulation Model (AUSM). The values of the exogenous parameters reflect views of the future that were appropriate in 1980. Nevertheless, the results presented here are the best projections currently available, and improvements must await development of the AUSM.

The other two strategies—STF and DAM—are analyzed in a recent report, together with the other major acid rain control bills that were introduced into the 97th Congress: Mitchell (S. 1706), Moynihan (S. 1709), and Moffett (H.R. 4829) (*6*). It is difficult to project with any accuracy the costs of implementing each of these bills because of the many options they give the states for achieving the required emission reductions. In this study, therefore, only approximate cost estimates for the bills are provided, with no attempt to project secondary impacts. The Congressional Research Service has summarized analyses of the acid rain control bills that have been performed to date (*7*). The cost estimates in these analyses are in general agreement with those presented here.

The analysis in this article is restricted to the so-called acid rain mitigation study (ARMS) region, which covers the 31 states east of and bordering the Mississippi River and the District of Columbia (see, for example, Figure 1). This is the region addressed in the bills considered here and commonly used in technical analyses of the acid rain issue.

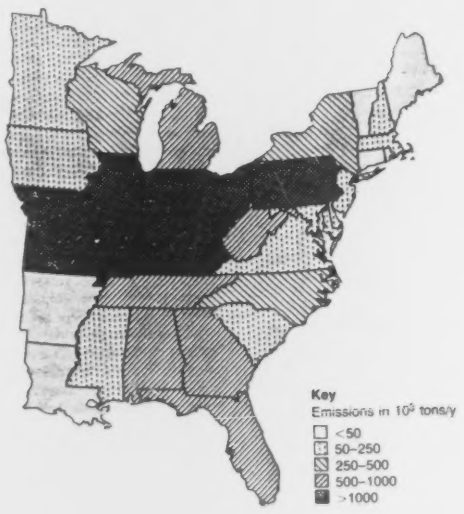
After discussing the regional costs

and emission reductions associated with each strategy, this article goes on to examine what effect each has on the following parameters:

- state-level emission reductions and costs,
- regional electricity prices,
- regional coal markets, and
- sulfur deposition.

The reductions in sulfur deposition resulting from emission changes are modeled using the advanced statistical trajectory regional air pollution (ASTRAP) model (*8*). Finally, the implications of the various strategies are discussed and compared with other approaches to reducing the potentially adverse effects of acidic deposition.

FIGURE 1
State-level distribution of 1980 utility emissions in the ARMS region



Specifications of each strategy

B95. The 1980 base case (B80) is compiled from a unit-level inventory created from DOE and EPA sources (*9*). Utility emissions are calculated from data on actual fuel use in the DOE/Federal Energy Regulatory Commission Form 67. The 1995 base case (B95) projects the emissions from utility sources in 1995 in the absence of any acid deposition control legislation. It includes new growth and retirement of older units and assumes that compliance with state implementation plans (SIPs) is achieved by 1985. Older units that were on-line prior to 1950 and smaller units with a generating capacity <100 MW are not required to meet the more stringent SIPs. In a sense, B95 represents a do-nothing strategy.

The emission reduction strategies are defined as follows:

CWM. This hypothetical approach consists of a mandatory requirement that all coal with sulfur content greater than 1.5% be cleaned by physical methods (*1*). The level of physical coal cleaning assumed in this analysis involves crushing the coal to a maximum diameter of 1.5 in. and flotation in a solution with a specific gravity of 1.6. The potential SO_2 emissions from the cleaned coal must be at least 10% lower than those from the raw coal.

CP2. This is the same as B95 except that a ceiling of 2 lb $SO_2/10^6$ Btu is placed on the allowable emissions from units meeting SIP limits. The advantage of such a ceiling is that emission reductions can generally be achieved by the use of low-sulfur or cleaned coal alone, without retrofitting the units with flue gas desulfurization systems.

STF. This strategy consists of implementing the Stafford bill, S. 3041 (a modified version of the Mitchell bill, S. 1706), approved by the Senate Committee on Environment and Public Works in July 1982 and introduced into the 97th Congress in November 1982. The Stafford bill sets a target SO_2 reduction of 8 million tons/y to be achieved in the ARMS region within 12 years of enactment. The decrease amounts to approximately 50% of the SO_2 emissions from the utilities in the 31-state region and 35% of the total SO_2 emissions in the region. State shares of the required reduction are to be determined in proportion to the ratio of the state's actual utility emissions in excess of 1.5 lb $SO_2/10^6$ Btu to the actual utility emissions in excess of 1.5 lb $SO_2/10^6$ Btu in the ARMS region. In addition, further reductions in SO_2 emissions from existing sources must be achieved to offset growth in SO_2 emissions from new sources. The growth offset has been analyzed elsewhere and is not addressed here (*6*).

The rationale behind the Stafford bill is to mandate a total level of reduction and then allow the states the flexibility of choosing their own methods of achieving it. Regional trading, energy conservation, early retirement, and least emissions dispatching are all permissible methods.

DAM. This approach entails implementation of the D'Amours bill, H.R. 4816, which was introduced into the 97th Congress in October 1981. The D'Amours bill is considered here because it attempts to focus the control strategy on the reductions that could be achieved by retrofitting flue gas desulfurization units on the power plants with the largest SO_2 emissions. The bill calls for a reduction equivalent to an 85% decrease in the SO_2 emissions from the 50 largest SO_2 emitters in the ARMS region. In addition, a ceiling of 1.2 lb $SO_2/10^6$ Btu is placed on the emissions from all other SIP units larger than 100 MW. Although the bill addresses the 50 largest power plant emitters in calculating the *magnitude* of the reduction, the *source* of the reduction can be any stationary source of SO_2 in the region. Alternative methods are allowed as under the Stafford bill.

It can be seen that these strategies

represent a range of possible approaches to the acid rain problem. The remainder of this article examines the relative merits of these strategies, assuming implementation by the year 1995. It is difficult to ascertain the uncertainties of the results presented here. To the best of our knowledge, no formal uncertainty analysis has yet been performed for the USM. Analysis of the two congressional bills is greatly hindered by the fact that it is impossible to predict which of several methods of reducing emissions or combinations thereof would be adopted by the states. The results should, therefore, be viewed as indicators of the relative effects of alternative strategies and not as accurate projections of the future. From our perspective in 1983, we believe that the growth in electricity demand used in the USM represents an unrealistically high forecast.

Since this analysis was performed, the acid rain issue has been taken up by the 98th Congress. In March 1983, Senator Stafford introduced S. 768, which is identical to S. 3041, the bill analyzed here. Simultaneously, he introduced a more restrictive bill, S. 769, mandating a 12 million tons/y reduction with offsets for new growth in SO_2 and NO_x emissions. Notable among other initiatives of the 98th Congress is the proposal by Representatives Sikorski, Waxman, and Gregg (H.R. 3400) to achieve a total reduction of about 14 million tons/y by retrofitting scrubbers on the 50 power plants with the highest SO_2 emissions and instituting other measures to decrease SO_2 and NO_x emissions from stationary and mobile sources. This bill is in many ways similar to the D'Amours bill. Work is under way to analyze these latest proposals in detail.

Regional impacts

In 1980, the actual SO_2 emissions from electric utility power plants in the ARMS region amounted to 15.9 million English short tons, most of which was from coal-fired units (*6*). This can be compared with an estimated 21.8 million tons/y from all sources in the ARMS region and 26.5 million tons/y from all sources of SO_2 nationwide (*10*). It is clear, based on economic, geographic, and feasibility criteria, that coal-fired power plants in the ARMS region represent the most likely target for major SO_2 emission reduction strategies. Figure 1 shows the state-level distribution of SO_2 emissions from utilities in the ARMS region in 1980. Table 1 summarizes the regional effects of the five control strategies.

Compared to the level of emissions in B80, the do-nothing strategy (B95) results in a modest decrease in annual emissions of approximately 0.3 million tons at no added cost. New growth in SO_2 emissions from stationary sources in the region is estimated to be about 1.9 million tons/y; a reduction of about 1.1 million tons/y would result from compliance with SIPs (*6*). The small decrease in emissions is a combination of these two factors, together with decreases resulting from the projected retirement of a number of older coal-fired units by 1995.

The coal-washing strategy (CWM) results in an overall annual reduction in emissions, relative to B80, of 1.7 million tons at a cost of approximately $820 million/y by 1995. At $470 per ton of SO_2 removed, this strategy is not very cost-effective. Also, it is unlikely that the limited SO_2 reductions obtained with this method would be large enough to substantially affect the amount of sulfur deposition.

The imposition of a 2 lb $SO_2/10^6$ Btu ceiling on allowable SO_2 emissions (CP2) yields an annual reduction of about 5.7 million tons at a cost of about $1.7 billion/y. This strategy costs $300/ton of SO_2 removed and is therefore more cost-effective than coal washing.

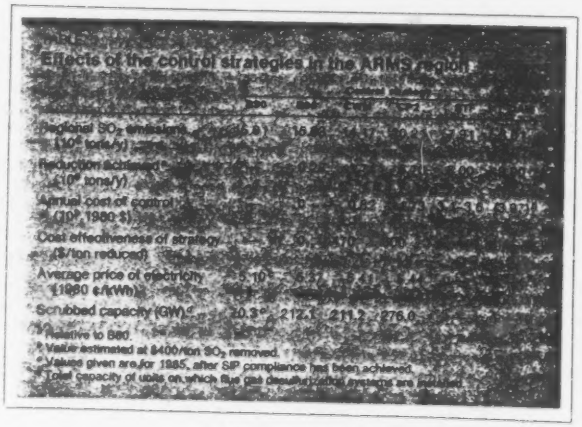
The two bills (STF and DAM) yield larger annual emission reductions of 8 and 9.9 million tons, respectively. Costs are difficult to determine for these two strategies because of the range of methods that can be used to implement them. The USM has not been used to project the effects of these bills. Cost estimates for STF performed by other groups are $3.1 billion/y (*11*) and $3.6 billion/y (*12*) in 1980 dollars. The D'Amours bill has not been analyzed elsewhere, but its cost is estimated at approximately $4.0 billion/y (*6*), using a value of $400/ton of SO_2 removed.

Under current regulations, the average price of electricity in the ARMS region is projected to rise from 5.1¢/kWh in 1985 (after SIP compliance has been achieved) to 5.37¢/kWh in 1995 (1980 dollars in both cases). The effect of both CWM and CP2 is to increase average prices by an additional 1% by 1995. The effects of STF and DAM would be more significant. ICF, Inc., estimates an increase by 1995 of 2.9% under STF (*11*).

The USM predicts that the generation capacity of power plants with scrubbers will increase from 70 GW (gigawatts) in 1985 to approximately 210 GW in 1995, with or without a coal-washing program (that is, with either B95 or CWM). Under CP2, about 65 GW of additional capacity would be scrubbed. Recent estimates of additional scrubbed capacity under STF vary widely from only 7 GW (*11*), to 22 GW (*12*), to a high of 89 GW (*13*).

Emission reductions and costs

Because sulfur dioxide emissions are currently greatest in the industrialized Midwest (Figure 1), the midwestern states would bear the brunt of the reduction requirement. Unfortunately, these same midwestern states are currently experiencing the worst of the economic recession and are consequently the least able to absorb any additional costs of pollution control. Table 2 lists the emission reductions


Effects of the control strategies in the ARMS region
Regional SO_2 emissions (10^6 tons/y)
Reduction achieved (10^6 tons/y)
Annual cost of control (10^9, 1980 $)
Cost effectiveness of strategy ($/ton reduced)
Average price of electricity (1980 ¢/kWh)
Scrubbed capacity (GW)
Relative to B80
* Value estimated at $400/ton SO_2 removed.
† Values given are for 1985, after SIP compliance has been achieved.
‡ Total capacity of units on which flue gas desulfurization systems are

required under each strategy in Pennsylvania, in the four midwestern states with the highest current emissions, and, for comparison, the state of Florida.

In the absence of acid rain control initiatives, emissions in 1995 are projected to be greater than 1980 emissions in three of these six states. The strategies discussed here require emission reductions ranging as high as 80% of the current utility emissions in Missouri. Clearly, under DAM the majority of larger power plants would be forced to retr- t a flue gas desulfurization sys- .n contrast, emission reductions .equired in Florida are no higher than 44%, due to the lower emission rates in that state.

Table 3 indicates that by 1995 control costs may increase by as much as $660 million/y for the state of Ohio. In contrast, a maximum increase of $130 million/y is projected for Florida. Cost-effectiveness is greater in the midwestern states, however, because a portion of the reduction in emissions can be obtained by switching fuels, cleaning coals, or blending coals. These options, however, entail disruption of traditional coal markets, which may have important social and political ramifications (discussed later in this article). Generally, the most cost-effective option is CP2, and the least cost-effective option is CWM. Figure 2 shows the pattern of state-level reductions in the ARMS region required by the most stringent control strategy: implementation of the D'Amours bill. It illustrates clearly that this stringent control program would have a potentially large impact on the industrialized midwestern states.

Regional electricity prices

The USM has a detailed financial module that simulates the costs and revenue requirements of 86 hypothetical utility firms, obtained by treating all investor-owned utilities within a given state as a single aggregate utility, and similarly treating all municipally owned utilities (*1*). It contains an accounting structure that provides an annual income statement, balance sheet, and sources and uses of funds for each of these "state" firms. The module also projects year-by-year utility expenses, including capital, operation and maintenance, and fuel. This structure enables the USM to project changes in electricity prices as a function of the implementation of a control strategy, where the price of electricity is defined as the total utility revenue requirements divided by the total sales to consumers. In this article, the effect of a single control strategy, CP2, on the price of electricity in the ARMS region is examined. The effects of STF and DAM are anticipated to be greater.

The regional average price of electricity in 1985 is projected to be 5.10¢/kWh. In the absence of any acid rain control program, this price would increase to 5.37¢/kWh by 1995—a 5.3% increase over the 1985 price. (All prices are in constant 1980 dollars.) Imposition of a 2 lb $SO_2/10^6$ Btu ceiling on allowable emissions would further increase this price to 5.44¢/kWh by 1995—a 1.3% increase over the 1995 price otherwise expected.

The effect on individual states in the region is variable, as Figure 3 shows. On the one hand, electricity prices in Ohio would increase by 16.8% between 1985 and 1995 under the current regulatory framework and by 21.4% with CP2. On the other hand, prices in Florida would increase by 4.1% under present regulations and by 4.7% with CP2.

Against this must be balanced the fact that electricity prices in the Midwest today are significantly lower than prices in the Atlantic Coast states. (The 1985 electricity price projection for Ohio is 4.1¢/kWh and for Florida is 6.4¢/kWh.) The proponents of strict control measures maintain that consumers in the East are currently paying higher prices for electricity partly because of a strong environmental stance taken in earlier years that included stringent SIPs and switching to clean fuels. Therefore, the increases in electricity prices in the Midwest must be accepted, they would argue, to bring midwestern states in line with the rest of the country. The Midwest would prefer an approach by which all states would contribute in appropriate, as yet undefined, amounts to the total costs of additional pollution control. The Sikorski-Waxman bill includes such an approach.

Regional coal markets

The effect of a particular control strategy on patterns of coal use can be important for determining the political viability of the strategy. The potential for major shifts in regional coal mar-

TABLE 3
Sulfur dioxide emissions under different strategies from utilities in selected industrialized states of the Midwest and Pennsylvania

State	Base	CWM	CP2	STF	DAM	Range of reductions[a] (%)
Ohio	2240	1803	1465	1400	1110	20–74
Indiana	1522	1470	1311	1061	1021	3–85
Pennsylvania	1488	1243	1243	1243	1243	0–51
Missouri	1131	1131	1131	1131	1131	0–80
Illinois	1110	1110	1110	1110	1110	0–80
Florida	1000	1000	1000	1000	1000	0–44
Rest of ARMS region	6889	6889	6889	6889	6889	0–80

[a] Range of reductions for the four control strategies, relative to base.

TABLE 4
Cost impacts of the four control strategies CWM, CP2, STF, and DAM on selected industrialized states of the Midwest and Pennsylvania

State	Range of energy cost increases (10^9/y)[a]	Range of coal price increases ($/ton)[b]
Ohio	180–660	400–920
Indiana	80–400	290–600
Pennsylvania	100–390	330–940
Missouri	30–360	220–610
Illinois	80–300	310–510
Florida[c]	30–130	400–580

[a] All costs in 1980 dollars.
[b] Covering the four control strategies, relative to base.
[c] Southern state included for comparison purposes.

FIGURE 2
State-level distribution of emission reductions required by the DAM strategy

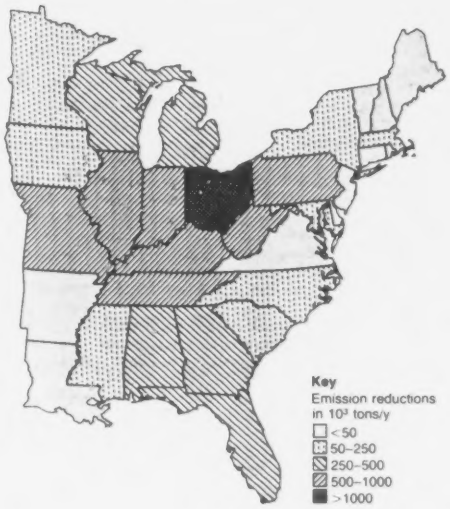
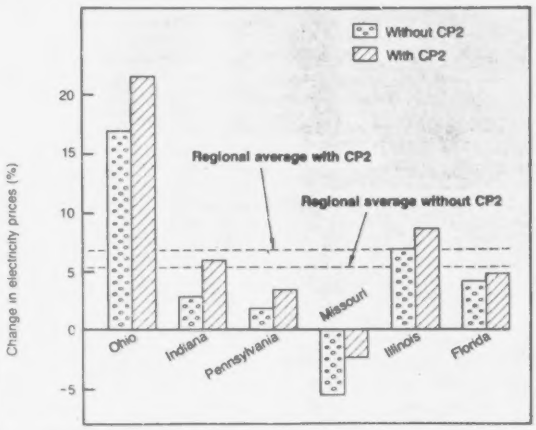


FIGURE 3
Changes in electricity prices in selected states in the ARMS region over the period 1985-95 under the CP2 strategy



kets can induce vigorous opposition to new initiatives in the states affected, which in turn can lead to powerful resistance in congressional committees and Congress itself. The protection of local coal industry production and jobs is a strong political force.

Implementation of emission reduction programs can force utility owners to switch fuels in order to comply with regulations at minimum cost. This may act to the detriment of traditional, local coal sources. Figure 4 shows projected patterns of utility coal consumption in the six states discussed previously, for the year 1995. For each state, the first bar represents the B95 strategy and the second bar represents the control strategy, CP2. B95 was chosen in preference to B80 in order to eliminate confounding factors arising from growth in coal use (that is, in Figure 4, the total coal consumption is approximately the same for the two strategies). CP2 was selected because it is the control scenario with the greatest potential for coal market disruption. This is because under a ceiling of 2 lb $SO_2/10^6$ Btu the optimum (least-cost) compliance strategy for the majority of plants would involve switching to a low-sulfur coal or blending an existing coal with lower-sulfur coals.

The more restrictive strategies, particularly DAM, are so stringent that flue gas desulfurization would probably be a requirement for many plants irrespective of coal sulfur content. Local midwestern coals, with high sulfur content, would therefore be preferred because of their lower cost.

Figure 4 reveals several interesting things. First, several states, exemplified by Pennsylvania and Florida, are relatively immune to coal market shifts, either because they have in-state low-sulfur coal supplies or because they are too far removed from alternative coal sources. It is interesting to find, however, that under CP2 in 1995, a certain amount of Powder River coal from Montana is projected to penetrate the Florida market, in spite of the large transportation costs involved. Second, several states in the center of the nation, typified by Missouri, currently use a number of different coal sources and are therefore quite flexible when control programs involving changes in emission limits are introduced.

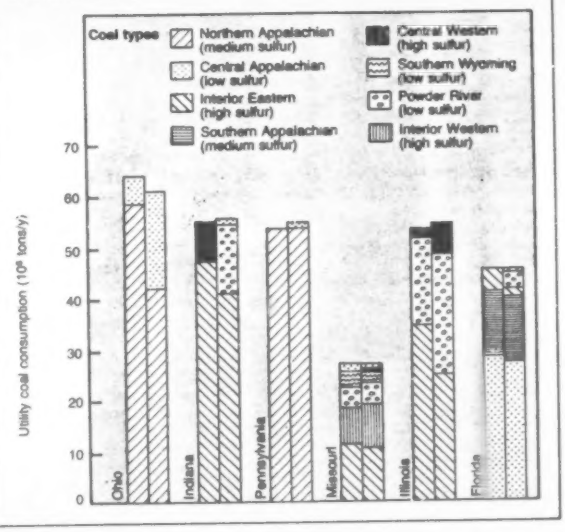
The states most vulnerable to coal market disruptions are those midwestern states such as Ohio, Indiana, and Illinois that currently rely heavily on relatively high-sulfur coals from in-state or from neighboring states. If a ceiling on emissions were imposed,

low-sulfur coals from Central Appalachia (in the case of Ohio) and from the Powder River basin (in the cases of Indiana and Illinois) would penetrate the local markets in significant amounts to be used as compliance coals. The projected reductions in local coal use are 28% for Ohio, 28% for Illinois, and 13% for Indiana in 1995.

Therefore, control strategies such as the ones described here clearly present a conflict. The most cost-effective strategies cause the greatest disruption in coal markets, whereas the least cost-effective strategies tend to preserve local coal interests (see box).

To examine this phenomenon further, USM was used to project the effects of these same strategies with protection of local coal interests (*1*). A strategy was developed, CL2, which is identical to CP2 except that coal-switching options are limited to locally produced coals, for those states that are themselves major coal producers. Plants in the affected states were permitted only to switch to local, in-state coals or to install flue gas desulfurization systems. The effect of this constraint was to increase regional costs in 1995 by 6% ($1.84 billion/y as opposed to $1.73 billion/y) for the same reduction in emissions (5.7 million tons of SO_2/y). States potentially benefiting the most from local-coal protection provisions are Illinois, Indiana, and Ohio.

FIGURE 4
Patterns of utility coal consumption in selected states in the ARMS region in 1995, by supply region, for the strategies B95 (first bar) and CP2 (second bar)



Sulfur deposition

The ASTRAP model was used to convert the changes in SO_2 emissions that would be produced by these control strategies into projected changes in sulfur deposition patterns (*8*). ASTRAP and almost all other operational models of long-range transport and deposition assume a linear relationship between emissions and deposition for any single source and specified meteorological period; that is, if the emissions from source A are reduced by one-third, the deposition at receptor B associated with emissions from source A will be reduced by exactly one-third. This linearity assumption makes the deposition from different sources additive and allows the models to be used repeatedly in examining alternative strategies.

However, if source-receptor relationships are not linear, as some experts hold, the value of such simulations may be limited. Certainly, the relationship must be linear if integrated over sufficiently broad scales (e.g., globally and annually). The recent acid rain report prepared by the National Academy of Sciences concluded that there is no convincing evidence of significant nonlinearity in eastern North America if averaged on an annual basis over spatial scales of 1000 km or more (*14*).

It is quite likely that some transformation and removal processes are nonlinear, as they would be if NO_2 and SO_2 are in competition for available oxidants. The crux of the linearity-nonlinearity question is whether marked nonlinearities exist over scales commensurate with either emissions decisions (on a statewide annual basis) or ecological, material, or health effects. (These effects are not examined here, but the most widely accepted view at this time is that they are a function of long-term total sulfur and nitrogen deposition, perhaps independent of speciation [*15*].) The answer is not yet known.

In addition to questions about the linearity assumption, general uncertainties in modeling of transport and deposition are ill-defined. Model sensitivity is an inadequate substitute for model uncertainty. The horizontal net mass flux of pollutants is not monitored, and dry deposition cannot yet be successfully monitored on a routine basis (*16*). Thus, in any model two of the three sink terms for pollutants cannot be compared with observed data. In addition, wet deposition measurements integrate the contribution of all sources and do not directly measure the effect of a single source.

In preliminary comparisons with measurements of annual wet deposition of sulfur in four U.S. and Canadian networks, ASTRAP simulations explained 75% of the observed variance in 1980. This is not offered as proof that ASTRAP correctly simulates the sulfur deposition changes that would be associated with these proposed emission changes, but it is encouraging. These simulations do not consider changes in emissions from nonutility or Canadian sources, nor emissions of other acidifying pollutants, oxidizing agents, or neutralizing agents, because that is beyond the scope of the proposed legislation examined here, and in part beyond the capability of the model.

For this exercise, it was considered appropriate to examine the changes in deposition associated with changes in utility emissions resulting from all U.S. and Canadian anthropogenic sulfur sources. All simulations are for one three-month summer season (June-August, 1980).

It is recognized that deposition

varies considerably from year to year. Indeed, wet deposition of pollutants is much reduced when precipitation itself is decreased, as in a drought, for example. To some extent, dry deposition then increases since more pollutant is available for this form of deposition. The meteorological data—primarily upper air winds and precipitation observations—necessary for many models exist. However, the time-consuming job of having a computer reformat the meteorological data into a usable synoptic form has not yet been undertaken. Therefore, to describe a systematic approach, a single season is examined here.

All simulations are for total (wet plus dry) deposition of anthropogenic sulfur. The 1980 base case in Figure 5 is calculated from an inventory of 27.9 million tons of SO_x emissions (expressed as SO_2) for the U.S. and 4.5 million tons for Canada. This inventory is a minor modification of the 1978 emissions inventory produced by the working groups established under the U.S.–Canadian Memorandum of Intent on Transboundary Air Pollution, and has seasonal and about 100-km resolution (*10*). The meteorological data—gridded 1000-mb and 850-mb wind fields and 24-h precipitation fields—were obtained from the Atmospheric Environment Service of Canada. The version of the model used here differs from that used in a recent report primarily because the wet removal parameters were adjusted according to a different resolution in the input precipitation data (*17*).

In the simulation using current total anthropogenic emissions, the maximum total sulfur seasonal deposition occurs in the upper Ohio River basin (Figure 5). It is in excess of 10 kg sulfur/ha. This simulation shows that almost all of the ARMS region and most of southeastern Canada have deposition rates greater than 2 kg sulfur/ha.

Table 4 shows how the sulfur deposition as calculated with the ASTRAP model (in kilotons of sulfur for the three-month period) varies with receptor region. The model predicts that approximately 60% of the total emissions are deposited on the North American continent. About 64% of that deposition occurs in the eastern U.S., and 17% occurs in eastern Canada.

The changes in deposition brought about by implementation of the five control strategies (B95, CWM, CP2, STF, and DAM) are simulated in the ASTRAP model by appropriate emission reductions at the state level applied to the utility sector component of the total inventory of SO_2 sources. The minimum resolution of the 1995 simulations is therefore statewide.

Figures 6–10 show the reductions in sulfur deposition achieved for each of the emission reduction strategies examined. Isopleths are of changes in deposition, measured in kg of sulfur/ha for the same three-month period. Note that Table 4 gives the same information according to region and control strategy. Changes in nonutility emissions and Canadian emissions are not considered.

It is interesting to observe that in the do-nothing strategy (B95), deposition is projected to increase slightly in two regions (eastern Canada and the western U.S.), and total deposition is reduced slightly. At the extreme opposite end of the range, implementation of the D'Amours bill (DAM) is estimated to reduce sulfur deposition in eastern North America by 41%—a reduction of 33% in total North American deposition.

The deposition simulations shown in Figures 6–10 can be examined with respect to the possible effects of the strategies on deposition in an ecologi-

FIGURE 5
Current patterns of sulfur deposition in kg/ha in the eastern U.S. and Canada from all anthropogenic sources

FIGURE 6
Sulfur deposition reduction in kg/ha in 1995 under the B95 strategy

FIGURE 7
Sulfur deposition reduction in kg/ha in 1995 under the CWM strategy

FIGURE 9
Sulfur deposition reduction in kg/ha in 1995 under the STF strategy

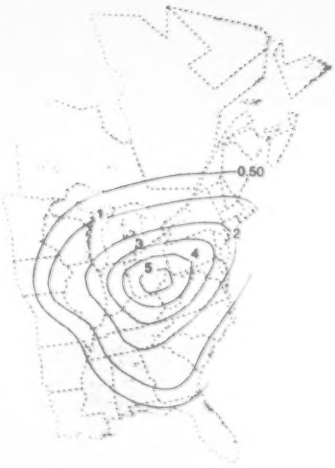


FIGURE 8
Sulfur deposition reduction in kg/ha in 1995 under the CP2 strategy

FIGURE 10
Sulfur deposition reduction in kg/ha in 1995 under the DAM strategy

cally sensitive region, the Adirondacks in New York State. According to the simulation of current conditions, the seasonal deposition of total sulfur here is about 3.8 kg sulfur ha^{-1}. Both B95 and CWM, which reduce emissions most in Tennessee, Kentucky, and Ohio, result in a deposition reduction pattern oriented southwest–northeast and reduce seasonal deposition in the Adirondacks by about 0.1 and 0.3 kg sulfur ha^{-1} $season^{-1}$, respectively. On the other hand, the emission reductions *in CP2, STF, and DAM significantly* affect many more states, and the resulting patterns of deposition change appear much like scaled versions of the current pattern (Figure 5), except in Canada and in parts of the Northeast where Canadian emissions contribute a significant share to the current pattern. The deposition reductions in the Adirondacks are about 0.8, 1.1, and 1.4 kg sulfur ha^{-1} $season^{-1}$, respectively, very nearly the same as the ratios of the overall emission reductions would indicate.

It is not surprising that in a linear model, broadly based emission reductions result in broad deposition reductions. At this time, proven methods are not available to translate these predicted changes in sulfur deposition to changes in measurable ecological response.

Discussion

The acidic deposition issue is fraught with major uncertainties. It is not known with certainty how much reduction in atmospheric emissions is necessary to mitigate the deleterious effects of acidic deposition, nor how that reduction should be assigned to achieve the greatest benefits for the least cost. And it is not known how quickly emissions must be lowered to protect sensitive receptors.

On the one hand, views have been expressed that the causal link between atmospheric emissions and adverse effects is tenuous, or, at best, unquantifiable. On the other, categorical assertions have been made that the link has been sufficiently demonstrated to support immediate action. Proponents of control have called for decreases in sulfur oxide emissions in excess of 10 million tons/y, while others recommend the treatment of selected lakes with lime and no reduction in emissions. It has been postulated that immediate action is necessary to prevent irreversible, long-term losses to sensitive ecosystems. The opposing viewpoint is that the time scale for damage is on the order of decades or longer and that the gradual replacement of old, uncontrolled plants by new, well-controlled plants is a sufficient strategy in itself.

This article has focused on a particular kind of strategy that would be favored if emission reductions are deemed necessary—a rollback in the emissions of SO_2 from coal-fired utility power plants in the eastern U.S. This kind of strategy has many advantages: The potential for the greatest absolute reduction exists in the utility sector in this region; it is almost certainly the *most cost-effective* strategy in terms of dollars per ton of pollutant removed; it is easy to implement; and it will have the minimum impact on the national economy per unit of pollution reduction. Arguably, it also addresses the actual cause of the problem, though the roles of local sources, oil-fired boilers, and non-SO_2 emissions are still being debated.

This article examines a range of SO_2 reduction strategies—a range both in terms of technical approach and in terms of the magnitude of SO_2 reduction (between 0 and 10 million tons/y). It does not advocate any particular approach because it is not known what magnitude and spatial configuration of reductions are necessary to alleviate adverse effects. Rather, an attempt is made to clarify the attributes of alternative types of strategies in order to facilitate the design of control programs that are efficient, equitable, and cause minimum disruption of other sectors of the economy. The "effectiveness" of a particular strategy is measured by the total amounts and regional patterns of changes in sulfur deposition as projected by the ASTRAP model.

Several conclusions regarding utility control programs can be made from this study. The do-nothing and mandatory coal washing strategies would in all likelihood be ineffective in reducing emissions and sulfur deposition by significant amounts by 1995. Coal washing is also less cost-effective than might have been anticipated. The effects of the Stafford and D'Amours bills would be very significant. Deposition would be reduced considerably, but costs would be high. Several midwestern states would receive economic shocks, with significant increases in electricity prices. An important observation on these strategies is that the deposition reductions are largest in the Ohio River basin and surrounding areas where present indications are that ecosystems are quite resilient to acidic deposition. The deposition reductions in areas with sensitive ecosystems such as the Adirondacks and Great Smoky Mountains are significantly less, drawing into question the effectiveness of such measures.

A 2 lb $SO_2/10^6$ Btu ceiling on SIP emissions would be cost-effective and produce reasonable reductions in deposition in comparison with the two bills. Its main drawback is that it causes disruption of existing coal supply patterns. One advantage of a simple ceiling on emissions allowed under SIPs is that regulatory enforcement and implementation of the program would be quite straightforward. The disadvantage of the flexible approaches such as STF and DAM is

TABLE 4
Changes in sulfur deposition as a result of implementing the control strategies

Receptor region	Current[a]	Deposition (10³ tons S/season)[b] ΔBase	ΔCWB	ΔCP2	ΔSTF	ΔDAM
Eastern U.S.	3467	40	143	413	545	681
Eastern Canada	398	6	10	67	85	100
Western U.S.	367	1	1	2	4	4
Western Canada	87	0	0	1	1	1
Total North America	2341	33	152	472	634	787
Percent of current eastern North American deposition	—	2	8	25	34	41
Percent of current total deposition	—	1	6	20	27	33
Emissions decrease[c]	3872[d]	35	219	717	1005	1243

[a] Deposition calculated by the ASTRAP model using meteorology for the three-month period June–August 1980. Units are English short tons of sulfur for the three-month period.
[b] Current deposition from all U.S.–Canadian man-made sources. Strategy Δ's are calculated relative to this base. A positive value indicates a reduction in deposition; a negative value indicates an increase.
[c] Decreases in emissions converted to tons of sulfur for the same three-month period.
[d] Current U.S.–Canadian anthropogenic emissions in the same units.

that a variety of technical solutions, including conservation programs and emissions trading, may be proposed that would frustrate implementation and enforcement activities.

The question of equity among the states applies to all the reduction strategies. Whether the costs of reducing emissions should be borne entirely by the states in which the largest emitters are located or whether all states in the ARMS region, or in the continental 48 states perhaps, should contribute in some way to the costs of control is debatable. One solution might be to impose a levy on all 48 continental states in proportion to, say, quantity of electricity produced (as in the Sikorski–Waxman bill) or quantity of SO_2 currently emitted. The fund generated could then pay for controls in the states where the most effective reduction could be achieved (see box).

Other approaches

How do these strategies to control SO_2 emissions from utilities compare with other possible approaches to the problem? Although the control of nitrogen oxide emissions from power plants would produce a reduction in acidic deposition, even less is known about the source–deposition relationship for nitrogen oxides than for sulfur oxides. Also, nitrogen oxides are generally believed to be less harmful to ecosystems than sulfur oxides. Several of the congressional bills (Mitchell, Moynihan, and Moffett) allow credits for reductions in NO_x emissions on the basis of two units of NO_x for each unit of SO_2 emissions, by weight. The potential for NO_x emission reductions in the utility sector is not high nor is it likely to be particularly effective. Utility NO_x emissions in the ARMS region are expected to rise from about five million tons/y in 1980 to six million tons/y in 1995 (*1*). However, the potential for reductions in NO_x emissions at existing boilers is limited. Improved combustion techniques such as low-NO_x burners, burners-out-of-service, and overfire air can reduce NO_x emission levels by only 30–50%, while the more efficient flue gas treatment processes are still under development and expected to be expensive.

ICF, Inc. has estimated that NO_x control strategies for utility boilers will be limited to about a 25% reduction in emissions, with cost effectiveness ranging from $50 per ton (for simple combustion modification of tangentially fired boilers) to $290 per ton (for wall-fired boilers) (*18*). The new limestone injection/multistage burner (LIMB) technology is promising, but not expected to be widely used before 1990.

Strategies that address sulfur oxide emissions from industrial fuel combustion or industrial processes, or nitrogen oxide emissions from transportation sources, will probably be ineffective in the absence of other measures. Industrial SO_2 emissions in 1995 are projected to be five million tons/y, and NO_x emissions from transportation are also projected to be about five million tons/y (*19*). In both cases, significant reductions in these levels would be cost-ineffective, difficult to implement and enforce, and potentially damaging to the economic well-being of specific industries. The utility sector is, in general, more robust in its ability to deal with price shocks than is the industrial sector, though it is certainly not as robust as it was in the past.

If it is determined that regional emission reductions are necessary or desirable, then it would seem reasonable to specify utility sources as the vehicle for reduction, or at least to calculate the required decrease on this basis, but to permit trading with other categories of sources at prescribed "exchange rates." If it is ultimately determined, however, that the goal of reducing acidic deposition is to protect relatively small receptor areas, and that local sources are the primary contributors to deposition, then the need for regional emission reductions might be obviated and specific emitters or specific states would be targeted for control irrespective of source category.

The possibility of an intermittent fuel switching strategy has been proposed (*20*) and its feasibility analyzed to a limited extent (*21*). The rationale for this approach is that the conversion of sulfur oxides to sulfate tends to be episodic, corresponding to periods of stagnating high pressure systems in the eastern U.S. during spring and summer. If a significant fraction of the total acidic deposition occurred during these relatively few episodes during the year, then intermittent fuel switching from coal to natural gas or other clean fuel during these periods might be effective in reducing total deposition.

The feasibility study of intermittent fuel switching does not support the effectiveness of such a strategy. Sulfate deposition was found to be somewhat episodic in the northeastern U.S., with typically about 30% of the annual deposition being contributed in eight episodes totaling about 50 days annually. However, a simulated reduction in SO_2 emissions of 50% at all coal-fired power plants in the upper Ohio River basin during a stagnation episode produced only a minor reduction in sulfur deposition in downwind areas. The technical and implementation problems of extending the reduction either in space, time, or magnitude further inhibit the attractiveness of the strategy. The need for meteorological forecasting, alert mechanisms, and compliance verification would create additional problems.

Finally, the possibility of "receptor mitigation," as opposed to "source

Update on pending acid rain legislation

Since this article was written, H.R. 4300 was introduced in the 98th Congress by Representatives Sikorski, Waxman, and Gregg. This bill requires a 10-million-ton reduction in SO_2 emissions and a 4-million-ton reduction in NO_x emissions. A large part of the SO_2 decrease is to be achieved by mandating that scrubbers be installed on the 50 power plants with the highest current SO_2 emissions. This requirement is included in the bill to minimize disruptions in the coal market (such as those discussed in this article). Because scrubbers are required on the 50 largest emitters, those plants would have no incentive to change to low-sulfur coal. Some smaller plants would probably switch fuels, causing certain changes in the coal markets but smaller disruptions than those projected under STF and DAM.

The Sikorski–Waxman bill establishes a 1 mill/kWh user fee on nonnuclear electricity generation throughout the 48 continental states. This money would be channeled into a trust fund to reimburse utilities for 90% of the capital cost of installing scrubbers. Thus, the cost of scrubbers, the major expense of acid rain control, would be spread throughout the nation. An analysis prepared by the Office of Technology Assessment projects that electricity rate increases under this bill would range from 1–10%, with an average of 3.2–3.7% during the most expensive years. The Sikorski–Waxman bill allows utilities less flexibility in choosing methods of SO_2 control than they would have under either STF or DAM, and the overall cost to the nation may be higher than that associated with either of the other two bills.

—Bette Hileman

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mitigation" should be considered. Liming of acidified lakes has been suggested as a solution to a problem that may be limited in geographical extent to a relatively few lakes in only a handful of areas. The obvious drawback to this approach is that it does nothing to ameliorate impacts on other ecosystems or materials, whether currently known, suspected, or unsuspected. There are several other disadvantages. Liming is not a permanent solution. Lime must be added to a lake periodically in an amount appropriate to the watershed and the current pH. Also, lime is a very impure substance that adds other pollutants such as heavy metals to the lakes.

Crocker et al. have attempted to evaluate the costs of a lake-liming strategy on the basis of several assumptions about the extent of liming necessary and the resultant lake pH levels that are desirable (*22*). The estimates range between about $400 million/y and $8 billion/y (in 1978 dollars). These costs are of the same order of magnitude as the costs of the SO_2 reduction strategies reported here. Fraser et al., in a study for the Electric Power Research Institute, estimate a cost of $100/acre for a lake-liming program, cautioning that the cost may vary significantly depending on the accessibility of the lake and its proximity to the limestone source (*23*). These investigators do not attempt to estimate the total costs of such a program. It is clear that further research is needed to fully identify the extent of liming that would be necessary and thereby to narrow the range of cost estimates.

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EPRI

AN ACCELERATED NATIONAL ACIDIC DEPOSITION RESEARCH AND DEVELOPMENT PROGRAM 1984-1988

AUGUST 9, 1983

ELECTRIC POWER RESEARCH INSTITUTE

Section 1

INTRODUCTION

The recent National Academy of Sciences report on Acidic Deposition, which examines only the atmospheric processes, prefaces its conclusions with the following two statements:

- "The central issues of concern in this report are the adequacy of current scientific understanding about the relationships between emissions and deposition, the extent to which the relationships are strongly nonlinear, and the extent to which distant sources contribute to deposition in ecologically sensitive, remote areas."

- "Much remains to be learned about the detailed mechanisms involved and their relative importance for the relationships between emissions and deposition. As scientists, our training leads us to be concerned about the current limits of our understanding of the relevant processes and the uncertainties associated with assessing cause and effect."

Other recent studies arrive at the same conclusion. Great shortcomings are evident in the available data. Limited knowledge exists on the transport of the precursors to acidic deposition, their physical and chemical transformation, and the processes by which the sulfates and nitrates are deposited.

Studies of the effects of acidic deposition find similar uncertainties on cause and effect. It is agreed that a number of lakes in sensitive areas have become acidified in the recent past, others have been acidic for a long time, and many remain neutral and have not changed recently. In many of the acidified lakes, fisheries are declining or have disappeared. It is also agreed that forest damage is taking place in certain areas of Germany and New England. Acidic deposition is hypothesized as one of many possible causes of this damage. It is generally

agreed that crop damage at current levels of deposition is small or nonexistent. Materials damage in polluted areas is taking place, but the contribution of acidic deposition is unknown.

In addition, more information is needed to apply the potentially most cost-effective approaches to reduce the emissions from power plants. Nevertheless, critical discussions are underway on the need for reducing the emissions from coal-burning power plants in the east and midwest. Currently, proposed regulatory and legislative actions to control the emissions of power plants in this region could affect some 200,000 Megawatts of generation. The costs of resulting plant modifications to achieve an 8 million ton per year SO_2 reduction have been estimated by the Edison Electric Institute to be as high as $15 billion. Other estimates based on less specific data have been lower, with the lowest at $2 billion.

The urgent need for a technically sound basis for evaluating the benefits and costs of reducing power plant emissions suggests the merits of a comprehensive R&D program on all aspects of acidic deposition. A national research program could provide scientific data to guide future decisionmakers on the needs and appropriate timing of emissions controls that should be established. Likewise, information on potentially attractive alternatives to current flue gas desulfurization practices could allow the selection of considerably more cost-effective options.

This document proposes a research and development program directed at achieving a substantial improvement in the scientific understanding of atmospheric pathways in the formation of sulfates and nitrates, the effects of acidic deposition, and the engineering development of cost-effective technologies to reduce the emissions from coal-fired power plants. The program represents a comprehensive extension of activities already underway at EPRI and within the federal agencies and has a high probability of providing better answers to the questions necessary to assess and deal with acid rain concerns in the next five years.

The prime objectives of this R&D would be to develop:

- A data base on the source of the precursors of the sulfates and nitrates in the atmosphere and the pathways to their deposition in critical areas.
- A physical and chemical understanding of the observed data and an analytical model that describes the key source-receptor relationships.
- An understanding of the ecological effects of acidic deposition on lakes, fisheries, forests, and crops.
- Potential methods to manage the effects of acidic deposition on the ecosystems.
- Most cost-effective methods of controlling power plant emissions that contribute to acidic deposition.

The proposed research and development divides into two general areas: Environmental Assessment to understand and predict the source, transport, and effects of power plant emissions and Environmental Control to develop cost-effective methods of controlling such emissions.

1.1 ENVIRONMENTAL ASSESSMENT

Environmental Assessment R&D addresses the physical and chemical processes that must be understood to explain the phenomena of acid precipitation and its effects. An outline of the intended work is shown below and more detailed descriptions can be found in Section 2.

Atmospheric Research

Perform experiments and analyses aimed at developing an improved theoretical understanding of the source receptor relationship and the analytical capability to predict the effects of emissions from specific power plants. Examples of the experimental research that would be conducted are:

- Large-scale tracer experiments (MATEX) using fluorocarbon tracers tc track plumes and air masses over the region east of the Mississippi.

- Cloud chemistry experiments to understand the reactions occurring between the precursor SO_x and NO_x molecules and the sulfate and nitrate deposition.

- Development of techniques to measure dry deposition and thereby understand its contribution to acidic deposition.

This experimental program would be accompanied by a theoretical and analytical program aimed at explaining the observed data.

Effects Research

Perform experiments aimed at developing an improved understanding of the ecological effects of acidic deposition and to learn ways to manage the effects on the ecosystems. Examples of key work that would be performed as part of effort are:

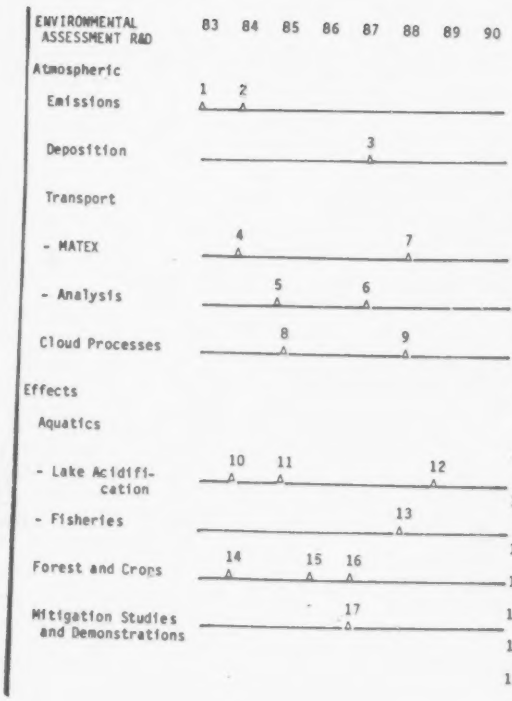
- Lake acidification experiments and validation of the understanding (ILWAS model) developed for the Adirondack Lakes for other parts of the country.

- Experiments on the effect of acidification on fish in lakes and streams. Develop a process model to describe these effects.

- Experiments on the effect of acidic deposition on tree growth and the development of a process model capable of analyzing the effect of acidic deposition on the health of a forest area.

- Experiments on the management of the effects of acidification of lakes (mitigation).

This work represents a significant acceleration over the current 1984-1988 EPRI plan. The timing of some of the key milestones are shown in Figure 1. The estimated cost of the effort on Environmental Assessment is $412 million over the next five years. Table 1 shows a distribution of the needs among the research areas. About $75 million of this research is already committed in EPRI's 1984-1988 plan. It is estimated that about



Representative Milestones

1. 1977 Eastern U.S. inventory available
2. 1982 U.S. inventory available
3. General Theory of Dry Measurement available
4. MATEX Tracer Experiment Feasibility established
5. 1st generation nonlinear model available
6. 2nd generation nonlinear model available
7. Empirical relationships established from MATEX data
8. Two Component Laboratory tests completed
9. Cloud studies data available
10. Lake Acidification Model - Validated Southern Appalachia Lake
11. Lake Acidification Model - Validated Midwest Lake
12. Generic Lake Acidification Model Validated
13. Generic Fisheries Model Validated
14. Forest Nutrient and Canopy Study Completed
15. Effects of Al on Trees Defined
16. Report Issued on Tree Damage from Acid Deposition
17. Initial Mitigation Study Results Available

MILESTONES ACCELERATED ENVIRONMENTAL ASSESSMENT PROGRAM

FIGURE 1

1984-1988 FUNDING REQUIREMENTS
($ MILLIONS)

	In EPRI 1984-1988 Plan	Estimated Federal Funding*	Requirements	
			Additional	Accelerated National Program
Atmospheric				
Emissions	0.1	14.9	0	15
Deposition (Wet & Dry)	4.8	20.2	5	30
Transport	24.7	15.3	110	150
Cloud Processes	10.9	14.1	35	60
Effects				
Aquatic	7.6	17.4	15	40
Forest	10.9	19.1	5	35
Crops	3.9	6.1	0	10
Materials	1.0	6.0	3	10
Health	0	2.0		2
Mitigation Studies	8.6	1.4	10	20
Mitigation Experiments	0	0	25	25
Assessment	1.9	10.1	3	15
TOTAL	74.4	126.6	211	412

TABLE 1

ENVIRONMENTAL ASSESSMENT 1984-1988 FUNDING REQUIREMENTS
ACCELERATED NATIONAL R&D PROGRAM ON ACIDIC DEPOSITION

*These are gross estimates based on current plans, and should not be construed as having the accuracy implied by three significant places.

$127 million is to be funded through the Federal Inter-Agency Acidic Deposition Research Plan. This would leave an additional unfunded requirement of a little over $210 million, nearly half of which is for MATEX.

1.2 ENVIRONMENTAL CONTROL

The proposed research and development on environmental control technologies focuses on the development of more cost-effective methods of reducing SO_2 and NO_x emissions than with current flue gas desulfurization systems. The brief outline of the intended effort is given below and more detailed descriptions can be found in Section 3.

Coal Quality Improvement
Experiments will be performed on advanced coal cleaning processes. Cleaning characteristics of regional coals will be defined.

Furnace Limestone Injection
Perform pilot-scale, prototype (20-100 MW) and demonstration (300-700 MW) tests of furnace limestone injection SO_2 control.

Combustion NO_x Retrofit
Full-scale demonstrations of retrofitted low-NO_x burners at three or more representative boilers.

Retrofittable Partial Scrubbing
Perform experiments, prototype tests (10-20 MW), and demonstration (100-200 MW) of retrofittable partial scrubbing processes.

Advanced Process Evaluation
Perform pilot-scale (1-2.5 MW) experiments and evaluations to establish the capability and economics of advanced flue gas desulfurization processes to achieve 50-90 percent SO_2 removal in a retrofit applications.

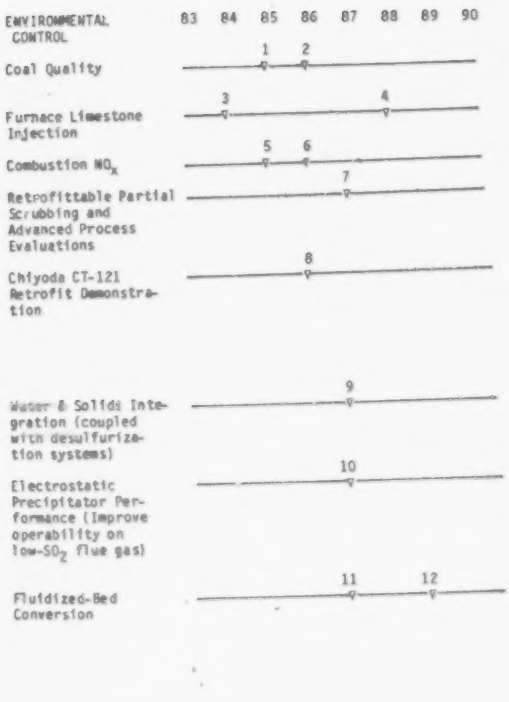
Chiyoda CT-121 FGD Retrofit Demonstration
Perform demonstration tests (150-200 MW).

Water and Solids Integration
Perform integrated water treatment and solids management experiments at one or more existing plants.

Electrostatic Precipitator
Experiments to upgrade precipitator performance for operation on low SO_2 flue gas.

Fluidized Bed Combustion
Demonstrate the retrofit of an Atmospheric Fluidized Bed to an existing 100 MW boiler.

Timing of some of the key milestones of the Accelerated Environmental Control Program is shown on Figure 2. Funding requirements for this effort are illustrated in Table 2. The estimated cost of the program is $480 million over the 1984-1988 periods. The EPRI Plan includes $95.5 million for work on the indicated subjects. An EPRI estimate of government funding for R&D on these technologies is about $40 million. Additional funding of about $345 million is required.



Representative Milestones

1. Complete characterization of 25-30 Bituminous Coals
2. Complete testing and guidelines for beneficiation and blending of low rank coals.
3. Complete pilot-scale development.
4. 20-100 MW prototype results complete. Early results from multiple full-scale demonstrations.
5. Complete test of prototype wall-fired burners.
6. Initial results from multiple full-scale tangential-fired and wall-fired burner demonstrations.
7. Results from in-duct spray drying and other partial SO_2 removal demonstrations.
8. Operating data available from 150-300 MW Demonstration.
9. Initial results from full-scale integrated system demonstrations.
10. Results available from full scale field evaluations.
11. Startup of retrofitted FBC boiler.
12. Complete performance testing of FBC Retrofit boiler. Design and operational data available.

MILESTONES ACCELERATED CONTROL TECHNOLOGY PROGRAM

FIGURE 2

1984-1988 FUNDING REQUIREMENTS
($ MILLIONS)

	In EPRI 1984-1988 Plan	Estimated Federal Funding	Requirements: Additional	Requirements: Accelerated National Program
ENVIRONMENTAL CONTROL				
Coal Quality Improvement	40			105
Furnace Limestone Injection	11			89
Combustion NO_x	7			36
Retrofittable Partial Scrubbing	5			60
Chiyoda CT-121 Retrofit	0.5			35
Advanced Process Evaluations	4			25
Water and Solids Integration	8			45
Electrostatic Precipitator Performance Improvement	5			25
Fluidized Bed Conversion	15			60
	95.5	40	345	480

TABLE 2

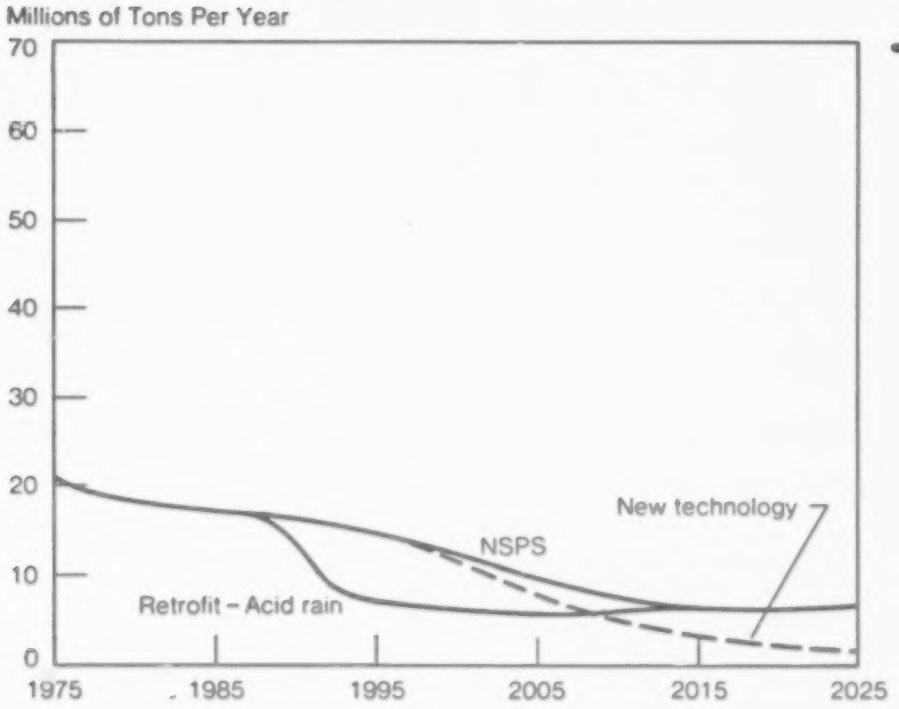
ENVIRONMENTAL CONTROL 1984-1988 FUNDING REQUIREMENT
ACCELERATED NATIONAL R&D PROGRAM ON ACIDIC DEPOSITION

1.3 OPTIONS FOR THE 1990s

The Electric Power Research Institute's research and development on Acidic Deposition is considered as a part of a broader program on environmentally acceptable use of coal for the generation of electricity. This broader program has enjoyed strong support since the formation of the Institute about 10 years ago. As a result, a substantial data base has been developed on the airborne, water, and solid effluents of power plants. Significant support has been given to the research on ecological effects of these effluents. A broad range of control technologies has also been tested to determine the effectiveness and economics of alternative approaches. The R&D being proposed in this document can make use of the foundation that has been laid by this earlier effort. For example, the data collected on Sulfate Regional Experiment (SURE) can now be the basis for developing an improved understanding and analytical models of the acidic deposition phenomena in the East. Likewise, key facilities such as Arapahoe Test Facility in Colorado and the Coal Cleaning Facility in Pennsylvania are available to do a number of the proposed projects.

A perspective on the effect of reductions of utility SO_x emissions can be gained from Figure 3. Utility SO_x emissions have been decreasing for some time. For example, the decrease since 1975 is shown. If one projected a hypothetical uncontrolled case, the SO_x emissions could be as high as 35 million tons per year in 2000. But new units would be expected to meet the NSPS standards, and hence the emissions should follow the NSPS curve as indicated. If an 8 million ton per year reduction, aimed at reducing acidic deposition, were to occur, the SO_x emissions could decrease as shown between 1985-1990. This level of emissions continues for one or two decades and then rises to meet the NSPS case as the new coal-fired plants are introduced.

UTILITY SO_x EMISSION TRENDS



Based on EPA emission data and projections adjusted to reflect current utility growth projections.

- Basis average coal sulfur content — 1.8%
- Retrofit control for acidic deposition reduced SO_2/yr by 8 millions between 1985–1990
- Advanced technologies phased in after 1995

Figure 3.

As new technologies, such as coal gasification combined-cycles and fluidized bed combustion, become demonstrated and commercialized, the total emissions could be reduced along the dotted pathway. This observation suggests the need for a balance between R&D support for technologies that can be adapted to retrofit applications and the new technologies that could be introduced in the latter part of this decade.

EPRI has, therefore, given substantial R&D support to the development of intermediate Btu coal gasification-combined cycle (GCC) and fluidized bed combustion (FBC) systems. Both of these options show the promise of reducing SO_2 and NO_x emissions to very low levels. These are likely to be the effective coal based choices for the new generation of electricity in the latter part of this decade.

The key coal gasification-combined cycle effort is the 100-MW Cool Water Project that is being demonstrated on the Southern California Edison system. This unit should be operating in 1984. This unit, located in California's Imperial Valley, will meet California's stringent emission standards for SO_2 and NO_x when using high sulfur midwestern coals. The level of interest in these new technologies is illustrated by the utilities' formation of the Utility Coal Gasification Associates (UCGA). The 24 members of this association represent utilities from all parts of the country. Their purpose is to encourage the development of coal gasification systems and to coordinate the exchange of information.

Research and development on Atmospheric Fluidized Bed Combustion has progressed to the stage of a 20 MW pilot plant now operating on the TVA systems. EPRI plans include the initiation of a 160 MW demonstration at TVA that should be in operation in 1989. The interest in this technology was demonstrated by the five excellent proposals that were submitted to EPRI.

Other options, such as the mix of nuclear power and coal units, could affect the total SO_x emissions. Coal gasification in conjunction with fuel cells offers other opportunities.

The EPRI R&D programs that address the coal-based alternatives are funded as shown below:

		($ MILLIONS) 1984-1988 Plan
o	Coal Gasification	76.6
o	Fluidized Bed Combustion	111.8
		188.4

The potential for a relatively rapid rate of introduction of these advanced coal-based technologies should be given careful consideration in the selection of a pathway toward reduced emission of the precursors of acidic deposition.

1.4 SUMMARY

A summary of the required support for the total Acidic Deposition Program is shown in Table 3. The total cost of the accelerated program is estimated to be $890 million. Government support is expected for a substantial part of the difference between the planned funding and that required for the accelerated effort. An EPRI estimate of the intended government support of specific R&D programs is subject to large uncertainties. Nevertheless, an estimate of about $167 million has been made based on current levels of support and our understanding of planned efforts. Over $550 million of additional funding is required. The subject of the source of this additional support should be addressed.

The total EPRI program related to the environmentally acceptable use of coal is substantially greater than the R&D on acidic deposition. When all of the aspects associated with the use of coal are included, the funding level shown in Table 4 results. This table includes the work on acidic deposition, other environmental

1984-1988 FUNDING REQUIREMENTS
($ MILLIONS)

	In EPRI 1984-1988 Plan	Estimated Federal Funding	Requirements Additional	Requirements Accelerated National Program
ENVIRONMENTAL ASSESSMENT				
Atmospheric Research	40.5	64.5	150	255
Effects Reaearch	32	52.0	58	142
Assessment	1.9	10.1	3	15
	74.4	126.6	211	412
ENVIRONMENTAL CONTROL	95.5	40	345	480
Total	$169.9	166.6	556	892

Table 3

SUMMARY CHART

ACCELERATED NATIONAL ACIDIC DEPOSITION R&D PROGRAM

PROGRAM	(MILLIONS OF DOLLARS) 1984	1984	1986	1987	1988	TOTAL
Advanced Power System						
Clean Gaseous Fuels	15.1	10.5	11.7	17.3	22.0	76.6
Coal Combustion Systems						
Air Quality Control	7.5	7.7	8.5	8.5	10.0	42.2
Desulfurization Processes	4.9	4.7	5.2	6.0	7.0	27.8
Integrated Environmental Control	2.3	2.5	3.3	6.0	5.0	19.1
Fluidized Combustion & Alternative Fuels	11.8	16.9	25.5	28.5	29.1	111.8
Coal Quality	7.2	7.5	7.8	11.0	12.0	45.5
Energy Analysis and Environment						
Environmental Physics and Chemistry	11.1	12.4	14.0	17.0	19.5	74.0
Ecolcgical Studies	8.0	9.0	8.8	11.8	14.2	51.8
Health Studies	2.4	2.3	2.8	3.2	3.8	14.5
	1.4	2.1	2.2	3.3	3.1	12.4
Economic and Environmental						
Integration	1.7	2.1	2.2	3.3	3.1	12.4
Annual Totals:	72.0	75.6	89.8	112.6	125.4	475.7

TABLE 4

CURRENT EPRI 1984-1988 PLAN R&D RELATED TO THE ENVIRONMENTALLY ACCEPTABLE USE OF COAL (DOES NOT INCLUDE ACCELERATED NATIONAL PROGRAM)

aspects of the use of coal, environmental control options, and the alternates being developed as options for new plants. The total support represents 28% of the funds available to EPRI during this period.

It is unlikely that this percentage could be increased without a serious effect on the other critical R&D being performed by the Institute. Support from other sources will be needed.

The Research and Development proposed in this plan is presented in the following three sections:

2.0 Environmental Assessment describing a comprehensive effort on atmospheric research, ecological effects, and economic and environmental integration.

3.0 Environmental Control Technology describing the proposed R&D on control technologies directed toward methods of reducing the emission of the precursors of sulfates and nitrates from existing power plants.

4.0 Options for the 1990's outlining the EPRI research and development on coal gasification combined cycle, fluidized bed combustion and other options for the cost-effective reduction of emissions.

Section 2

ENVIRONMENTAL ASSESSMENT R&D

The proposed Environmental Assessment research and development includes a comprehensive effort on atmospheric research, ecological effects, and environmental and economic integration. The primary objectives of EPRI's Acidic Deposition scientific research program are:

1. To elucidate whether damage is being caused and, if so, by what mechanism and at what rate,

2. To determine what actions can be taken to reduce such damage either by reduced deposition or management of the damaged resources,

3. To identify the relationship between emissions and deposition and to develop the predictive capability of the effect of an increase or decrease in emissions at a specific point on a specific deposition site,

4. To evaluate the cost of such damage and the benefit from reduced emissions or other mitigation measures.

The projected cost of the total program over the next five years is shown in Table 5 as $412 million. Currently planned EPRI programs include $74.4 million for this effort. Some of the key milestones are illustrated on Figure 1. (Section 1-Introduction)

	1984	1985	1986	1987	1988	TOTAL	1984-1988 Totals EPRI (planned)	Federal (est.)*	Additional Needs
1. ATMOSPHERIC									
Emissions	2.6	2.8	3.1	3.2	3.3	15.0	0.1	14.9	0
Deposition	6.7	7.3	7.2	4.7	4.1	30.0	4.8	20.2	5.0
Transport	10.3	23.4	36.9	48.4	31.0	150.0	24.7	15.3	110.0
Cloud Processes	10.5	12.3	14.7	12.9	9.6	60.0	10.9	14.1	35.0
2. EFFECTS									
Aquatic	8.8	10.0	9.4	6.9	4.9	40.0	7.6	17.4	15.0
Forest	5.0	6.1	7.5	9.2	7.2	35.0	10.9	19.1	5.0
Crops	2.8	2.7	2.0	1.4	1.1	10.0	3.9	6.1	0
Materials	1.8	1.8	2.4	2.0	2.0	10.0	3.9	6.0	3.0
Health	0	0.5	0.5	0.5	0.5	2.0	0	2.0	0
Mitigation Studies	2.6	4.1	5.0	4.1	4.2	20.0	8.6	1.4	10.0
Mitigation Demonstrations	0	3.0	6.0	8.0	8 0	25.0	0	0	25.0
3. ASSESSMENT	2.8	3.1	2.8	3.0	3.3	15.0	1.9	10.1	3.0
	53.9	77.1	97.5	104.3	79.2	412.0	74.4	126.6	211.0

ACIDIC DEPOSITION RESEARCH PLANNED FUNDING AND ADDITIONAL NEEDS
($ MILLION)

TABLE 5

*These are gross estimates based on current plans and should not be construed as having the accuracy implied by three significant places.

2.1 ATMOSPHERIC RESEARCH

2.1.1 Emissions

Issue. The concern over acidic deposition focuses mainly on sulfates and nitrates, the precursors of which are principally SO_2 and NO_x. In order to relate acidic deposition to emission, we need quantitative information on the sources of the precursors (as well as on the amount of deposition.) Specifically, we need data on the distribution and quantity of material being emitted, both naturally and anthropogenically.

Response.

A. Approach

1. EPRI

   a) Develop a research-type inventory for anthropogenic sources using NEDS and other data, the inventory to be based on an 80x80 km grid for the United States and Canada. Averaging is on a seasonal basis except that hourly data are developed for power plants during particular intensive research episodes.

   b) Measure natural sulfur emissions from selected sites in the eastern United States, and extrapolate on a regional basis.

2. Federal

   a) Develop a statewide inventory of anthropogenic sources from NEDS. Averaging tends to be on an annual basis, with seasonal updating.

   b) Measure natural emissions from both land and waters, including oceans.

B. Comment

The two inventories are similar in the sources and emissions being covered but the EPRI inventory is developed more for a research mode (greater detail and precision) than for a compliance mode. The spatial resolution of the two inventories is different. EPRI is no longer making any inventory of natural sources; the federal program involves a significant effort in that area.

Funding. (See Table 5) The present program for emissions inventories seems adequate; hence, we see no need for additional monies over the next 5 years.

Deliverables.

A. EPRI: Eastern U.S.A. inventory for 1977: available now.
Entire U.S.A, inventory for 1982: 1984

B. Federal: Inventory through 1980: available now
Inventory continuously being updated.

2.1.2 Deposition

Issue. An important national concern is that the deposition of the acidic material (principal sulfates and nitrates) is causing damage to the environment. In order to assess the damage, we need accurate information of the amount of material being deposited in both wet (precipitation) and dry forms.

Response.

A. Approach

1. EPRI

a) EPRI's approach to measuring wet deposition involves sampling on an event basis at 32 sites in the eastern third of the U.S. Samples are analyzed for acidity, sulfate, nitrate plus a number of other consituents.

b) The approach being developed by EPRI to measure dry deposition involves use of mathematical model coupled with data on concentration gradients of pollutants in the atmosphere. We know of no way to measure directly the deposition of dry materials, hence, the need for a predictive model based on air quality and surface characteristics of the objects receiving the deposition. EPRI also is investigating an isotopic approach to measuring deposition.

2. Federal

a) For wet deposition, the program focuses on event sampling at 8 sites in the East (MAP-3S), weekly collections at about 150 sites throughout the U.S. (NADP, NTN), and weekly collections at some foreign sites (GTN).

b) For dry deposition, an approach similar to and complementary with EPRI is being used.

B. Comment

EPRI has excellent coordination with the MAP-3S program because of the use of identical procedures, but at

different sites, hence a comprehensive picture of deposition for the East is developing. NADP/NTN use different procedures but by combining data from all programs, we will have a good knowledge of total deposition and trends. For dry deposition, the Federal and EPRI approaches are similar. Both use a modeling approach. EPRI has developed some models and has joined with the Federal Program in jointly funding field measurements. Coordination of the two programs is excellent.

Funding. (See Table 5) Considering the talent available for research, the present program is nearly at a maximum level. We see a need for another $5 million over the next five years, mainly for additional research on dry deposition. The response of a variety of surfaces to receiving dry material is needed. About $2 million is needed for some additional precipitation measuring sites.

Deliverables. Data on eastern wet deposition through 1982 are available now. A high-quality, 10-year data base on the East will be ready in 1988. Data from NADP and NTN are continually being reported. Models for dry deposition will be available about 1986; field applications by 1988.

2.1.3 Transport

Issue. A fundamental question deals with the relation between emissions and deposition. Most attempts to answer this question focus on use of mathematical models, an important component of which is the transport of pollutants over long distances. Until we improve our knowledge of the transport component, we are unable to develop models which give meaningful predictions of the effect of emissions on deposition.

Response.

A. Approach

1. EPRI

a) EPRI has focused on both field studies and model development. The principal field studies have been the Sulfate Regional Experiment (SURE), a study of transport over the North Sea, and the current Regional Air Quality Studies (RAQS). These studies provide data for model development. EPRI is now conducting a feasibility study for a large-scale tracer experiment (MATEX) with the hope of funding the experiment with the Federal government.

2. Federal

a) A small-scale tracer experiment is underway (CAPTEX-jointly funded with EPRI). Additional field studies, along the line of MATEX are being considered. Extensive development of nonlinear models is in place.

B. Comment

EPRI and federal coordination is excellent. We have joined forces on projects (CAPTEX tracer study) and on planning (MATEX). Similarly, modeling activities are closely coordinated.

Funding. (See Table 5) Despite the large amount of money in this area of research, we see a national need for another $110 million. This money is needed for a more definitive field study along the lines of MATEX - one that actually traces emissions from source to point of deposition and includes an evaluation of chemical transport. About $15 million of the $110 million is needed for specialized field studies and model improvement.

Deliverables. SURE data: available now. Evaluation of regional transport models will be completed in 1984, as will the MATEX feasibility study. The first generation of nonlinear models will be ready in 1985, the second generation in 1987. MATEX is scheduled to end in 1988. New, comprehensive models will be ready in 1989 or 1990.

2.1.4 Cloud Processes

Issue. The processes by which atmospheric gases or particles become incorporated into cloud water to become acidic precipitation are poorly understood. An understanding of such processes is essential to developing a model which can be used to predict deposition in terms of emissions. Research is needed, therefore, to understand the manner by which water actually becomes acidic.

Response.

A. Approach

1. EPRI

a) Laboratory research is aimed principally at chamber studies on processes that simulate cloud action. Present work is with simple 2-component systems. Multicomponent system research is planned.

b) Field studies focus on collecting samples under a variety of meteorological conditions - air samples collected near clouds, and water samples collected from the clouds.

c) The results of field and laboratory research are combined into a comprehensive picture.

2. Federal

a) Laboratory studies focus on specific reactions and kinetics.

b) Field studies, similar to those conducted by EPRI, are planned for the future.

B. Comment

EPRI and the federal government both use field and laboratory approaches. They differ in the laboratory reactions being investigated and in the geographic location at which field work will be done. No overlap exists.

Funding. (See Table 5) A large amount is already committed to this type of research. We see the need for an additional $35 million, about $30 million of which is for a field study of nonlinear reactions being proposed by the National Laboratories to the Interagency Task Force on Acidic Precipitation. An additional $5 million are needed for additional chamber research.

Deliverables. Results of laboratory study on simple 2-component systems will be completed in 1985. Results on more complex systems will not be available until 1988. Field studies on cloud dynamics will be completed during the period from 1988-1990.

2.2 EFFECTS RESEARCH

2.2.1 Aquatic Effects

Issue. Lakes have gone acidic and some lakes have lost their fisheries. Three questions remain unanswered: (1) the extent of lake acidification (in space and over time), (2) the relation between acidity and atmospheric deposition, and (3) the effect of lake acidity on biota. Answers to these questions are needed if we are to judge the effect of atmospheric deposition on aquatic ecosystems.

Response.

A. Approach

1. EPRI

a) With regard to causes of acidification, EPRI is focusing on processes, specifically, developing a mechanistic model which simulates the response of a lake to atmospheric deposition. The model has been developed for three lakes in the Adirondacks. It is now being tested in different parts of the United States.

b) EPRI is also conducting a paleolimnology study of 40 U.S. lakes (emphasis on diatoms) in order to date the time at which lakes became acidic.

c) EPRI fisheries research focuses on using a combination of laboratory and field studies to develop a model which predicts the fishery response to changing lake acidity.

2. Federal

a) An extensive program is underway to identify areas with waters susceptible to acidification.

b) A second effort involves obtaining baseline data on chemistry (including acidity) of selected lakes throughout the United States.

c) A monitoring program, over time, is also part of the program which includes biology as well as chemistry.

d) Population modeling based on field surveys is part of the fisheries research.

B. Comment

In general, the EPRI approach focuses on processes, the Federal on empirical field relations. The two, therefore, complement each other.

Funding. (See Table 5) An additional $15 million is needed in order to develop information which has broad application rather than gathering data from only a limited number of sites. Studies of watersheds in a number of different environments are needed if we are to develop predictive capabilities which apply in a general way under many different situations. The additional money would be used to study additional lakes (in different settings), to continue and expand the population studies so that more generic information could be developed; and to expand the bioassay work to cover more parameters than the very limited number now planned.

Deliverables. A 3-lake Adirondack acidification model is now available. Testing of the model in the Southern Appalachians will be completed in 1984, an upper Midwest model in 1985. A generic model may be available by 1989. A preliminary U.S. sensitivity map is now available. National and detailed regional maps will be ready in 1985. Monitoring data will be continually reported. A validated fisheries model will be available in 1988. Population modeling, based on field surveys, will be available after 1985.

2.2.2 Forest Effects

Issue. Acidic deposition has the potential for affecting forest productivity. Some areas receiving acid precipitation do show tree damage. The extent to which the acidity is responsible for the damage is uncertain. Other studies show that tree growth may be sensitive to toxicity from aluminum released from soil as a result of soil acidification. Questions remain about the extent of damage, the role of natural sources versus deposition acids, and the mechanisms by which productivity may be affected as a result of acidic deposition. Documentation of actual forest effects from acidic deposition remains to be demonstrated.

Response.

A. Approach

1. EPRI

a) Microcosms are used to study tree response to controlled conditions.

b) A second approach involves a study of canopy effects on acidic input and the effect of acidic input on nutrient cycling.

c) A major effort focuses on the mobilization of aluminum and its effect on productivity,

2. Federal

a) An extensive study of tree rings (from 70 forests in 14 states) as an indicator of growth is underway.

b) A study of North American red spruce dieback is in the plan.

c) Seedling response to acids is being investigated.

d) Future research will focus on aluminum mobility.

e) Future studies include role of acid deposition on predisposition of trees to diseases and pests.

B. Comment

EPRI and the federal government have different approaches. The federal approach focuses to a large extent on the response of the tree itself whereas EPRI research involves mainly a study of the effects of acids on processes which affect growth, viz, nutrient cycling. The federal program looks more at direct damage whereas EPRI studies more the processes leading to damage. The two complement each other.

Funding. (See Table 5) Between EPRI and the federal government, a well-designed research program is in effect. (A need, however, exists for an additional $5 million over the next five years to include additional sites, both in terms of type of trees and geographic location.)

Deliverables. The results of microcosm are available now. Nutrient and canopy studies will be completed in 1984. The results of the aluminum research will be available in 1986. A preliminary assessment of tree ring studies will be ready by late 1983. The seedling and red spruce studies will be completed after 1985. A fairly definitive report on tree damage from acid deposition could be available by 1987.

2.2.3 Crop Effects

Issue. Concern has been raised about the effect of acidic deposition on crop productivity. The economic implications of any damage are great. Although no damage to plants has been observed under present ambient conditions, research has been initiated to assess effects.

Response.

A. Approach

1. EPRI

a) Emphasis is principally on laboratory study of mechanisms affecting growth. Research involves plants from the Southeast, Northeast, Midwest, and Atlantic Seaboard.

2. Federal

a) Much research involves use of a dose-response approach on specific crops through controlled studies. Emphasis is shifting from laboratory to field studies.

b) A second federal approach looks at acidic deposition effects on soil processes.

B. Comment

EPRI emphasizes mechanisms affecting growth. The Federal program deals in dose-response. In the area of soil response, the two programs are somewhat similar except for the types of soils being studied. Because of the absence of evidence of effects under ambient conditions, EPRI is shifting its emphasis from acidic precipitation to mixed gaseous pollutants.

Funding. (See Table 5) We see no need for additional money in view of the programs in place and in view of the absence of any documented effects under present ambient conditions.

Deliverables. The results of EPRI's crop research will be available in 1984. The Federal studies on potatoes, clover, and fescue are now available. Results of other crop studies will be

available in the time period from 1985-1988. No date can yet be assigned to the availability of soil response results.

2.2.4 Materials

Issue. Acids are capable of having an adverse effect on materials (buildings, monuments, etc.), hence acid precipitation may be harmful to materials. The extent of damage cannot yet be ascertained. Furthermore, at present, we are unable to separate effects from acid deposition from effects due to air pollution in general. In assessing the cost to society from damage due to acidic deposition, we need information on the damage to materials, as well as to crops and ecosystems.

Response.

A. Approach

1. EPRI

a) EPRI has been involved only in studies which deal with estimating the economic cost of damage.

2. Federal

a) The federal program has undertaken a study of measuring damage in the field.

b) The program also has plans for a study of protective treatments.

B. Comment

EPRI is playing little role because of the federal activities. EPRI focuses, at present, only on cost estimates of damage. The federal research involves actual measurements of damage and will look into protective measures.

Funding. (See Table 5) We see a need for a small amount ($3 million) over the next five years to focus on two issues:

1. The need to separate damage from acidic deposition from that caused by air pollution in general, and

2. Investigation into protective schemes.

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Deliverables. The results of an EPRI feasibility study on economic costs will be available in late 1983. The final assessment will be delivered in 1985. The first results from the Federal field study (effects on tombstones) will be ready in late 1983. Other effects research will be reported during the period 1985-1988. No date has yet been assigned to completion of research on protective devices.

2.2.5 Health

Issue. Indirect health effects due to either accumulation of toxic chemicals in food chains or mobilization of metals, e.g., lead, into drinking water, especially in cases where precipitation is collected in metallic plumbing systems and used directly, without treatment. Such mechanisms have been postulated, but it is generally agreed that the risk is small.

Response.

A. Approach

1. EPRI

No work planned.

2. Federal

The federal government plans to investigate this issue only on a small scale. They will examine potential indirect health risks through documentation of potential exposures and risk assessment, and to examine feasibility of mitigative measures, if necessary.

Funding. (See Table 5) A minimal amount of research ($2 million) over the next five years should be sufficient to conduct exposure and risk assessments.

Deliverables. Exposure and risk assessments, 1988.

2.2.6 Mitigation Studies

Issue. If lakes have lost their fisheries or if forests have been affected by acidic deposition (or by other acidic inputs), proper management could improve the status of the lakes or forests. Management could include such practices as chemical treatment or selective stocking of fish. At present, we have some information on the effects of, for example, chemical treatment (liming) on lakes or forests. We know that liming would increase the pH of lakes but many additional effects can not yet be quantified. Research is needed to document the effects of mitigation on the chemistry of the system and on the biotic response.

Response.

A. Approach

1. EPRI

a) EPRI is starting a program of laboratory and controlled field studies to evaluate the effect of liming on chemistry of the system, mainly on lake systems.

b) The program underway will also involve a study of the effects of mitigation on biota.

2. Federal

a) The federal program is still at a nascent stage. Plans call for an assessment of available information and an approach to lake mitigation through laboratory and field studies. A 25-lake research/demonstration project is planned for late 1984.

B. Comment

The federal and EPRI approaches seem to be similar. Both are in the initial or conceptual stages, hence planning for a well-coordinated approach is feasible. EPRI is expanding its program to include a stronger biological component and to start an investigation into forests mitigation.

Funding. (See Table 5) Because the budget is modest now, we see a need for considerable monies over the next 5 years. We anticipate a need for an additional $10 million to cover more thorough research on lake mitigation, especially an investigation into the total biotic community. Furthermore, research on forest mitigation will require much more additional money because of the complexities of the forest system.

Deliverables. A preliminary evaluation of existing practices is available both from the federal and EPRI programs. No meaningful results from the field and laboratory programs can be expected before 1986.

2.2.7 Mitigation Demonstrations

Issue. Research underway and planned on mitigation will answer questions dealing with the techniques of mitigation and with the likely effects; however, the true effectiveness of mitigative measures cannot be judged without full-scale demonstrations. Some demonstrations have taken place in Europe but these have not involved a measurement of all the parameters we feel are important and they have involved lakes only under a limited set of watershed conditions.

Response.

A. Approach

No definite plans exist yet either by the federal government or by EPRI. If implemented, such a demonstrations program would involve individual lakes (and possibly streams), entire watersheds, and stands of forests. Demonstrations would take place under a variety of topographic, geologic, and climatic situations. Federal program may initiate a 25-lake demonstration project in late 1984.

Funding. (See Table 5) Nothing has yet been budgeted for this type of work. We anticipate that $25 million would be needed to carry out comprehensive and definitive demonstration projects.

2.2.8 Environmental/Economic Assessment

Issue. There is an important need for an acid deposition integrating framework for balancing the potential environmental effects with the costs of emissions control. Such a framework would bring together the many aspects of the acid deposition problem, ranging from control technologies and costs, transport and deposition, mitigation options, ecological and other effects, valuation of these effects, and research results. In addition, there is a need to develop methods to identify least cost management and emission reduction strategies for reducing damage to sensitive areas.

Response.

A. Approach

1. EPRI

EPRI has developed a computer model, the Acid Deposition Decision Tree (ADEPT) to integrate the various aspects of the acid deposition problem. In addition, EPRI has developed and begun to implement methodologies to value ecological and other damage costs. EPRI has also started to develop models which enable utilities to achieve cost-effective emission reduction strategies.

2. Federal

EPA has been funding the development of AUSM which is intended to estimate utility emissions under alternative control strategies. In addition, the federal government has initiated the development of a large-scale model designed to identify cost-effective national strategies for reducing deposition in sensitive areas. EPA efforts are also underway to develop and implement methods to evaluate ecological and other damage costs associated with acid deposition.

B. Comment

EPRI has excellent coordination with the federal program. EPRI personnel are used as reviewers for AUSM and the EPA integration effort. Although, in the past, EPRI efforts have predominantly focused on providing tools and information for the national debate, it is

anticipated that future EPRI efforts will emphasize the development of tools and information to enable utilities or groups of utilities to assess deposition reduction strategies and to determine cost-effective methods to reduce deposition in sensitive areas. Thus, the EPRI and EPA efforts are viewed as complementary.

Funding. (See Table 5) With EPRI's increased emphasis on providing utility specific and utility group integration tools, there is an additional need for three million dollars funding over five years.

Deliverables.

A. EPRI: ADEPT (Acid Deposition Decision Tree Model); available now.

Models for utility deposition reduction strategies: available 1986.

Valuation of environmental damage: available 1985.

B. Federal: AUSM: available 1984.

EPA's Acid Deposition Integration Model: available 1985.

Section 3

ENVIRONMENTAL CONTROL TECHNOLOGY

A limited R&D program on cost-effective technologies to control emissions from pulverized coal power plants is included in the EPRI 1984-1988 plan. This program requires a funding level of $95.5 million over the next five years. An accelerated effort is proposed that will supply the key information for a number of important options in a more timely manner. This accelerated program is outlined in Table 6. Descriptions of each of the intended efforts are included in this section. Key milestones are described in the text and shown in Figure 2. (Section 1.0 - Introduction) Funding requirements for this program total $480 million over a five-year period. Our understanding is that the government funding for similar programs is about $40 million.

TECHNICAL RESPONSES - SCHEDULE AND FUNDING REQUIREMENTS ($M)

Response	84	85	86	87	88	Total	Proposed 1984-1988 Plan
COAL QUALITY IMPROVEMENT							
Conventional Coal Cleaning	5	10	10	10		35	
Advanced Coal Cleaning	6	6	6	6	6	30	
Low Rank Coal Beneficiation	2	3	3	4	3	15	40.0
Coal Characterization	5	5	5	5	5	25	
FURNACE LIMESTONE INJECTION							
Prototype Tests (3)	6	8	6	2	1	23	
Full-Scale Demos. (2)	2	15	20	10	3	50	
Pilot Studies	6	4	3	2	1	16	11.0
COMBUSTION NO_x							
Full-Scale Demos. (3)	5	14	8	7	2	36	7.0
RETROFITTABLE PARTIAL SCRUBBING							
Prototype Tests	3	4	2	1		10	
Economic Tradeoffs	1	1	1	1	1	5	
Full-Scale Demo	5	15	15	5	5	45	5.0
CHIYODA CT-121 RETROFIT							
150 MW Demonstration	6	12	12	5		35	.5
ADVANCED PROCESS EVALUATIONS							
Pilot Evaluations	3	5	5	5	2	20	
Engineering/Economic Eval.	1	1	1	1	1	5	4.0
WATER & SOLIDS INTEGRATION							
Full-Scale Demos. (2)	5	10	15	10	5	45	8.0
ELECTROSTATIC PRECIPITATOR PERFORMANCE IMPROVEMENT							
Upgrade Technology	3	3	3			9	
Field Evaluation		2	2	7	5	16	5.0
FLUIDIZED BED CONVERSION							
100 MW Demonstration	20	30	5	3	2	60	15.0
Total	84	148	122	84	42	480	95.5

TABLE 6

Note: An EPRI Estimate of the Government Support of Similar Programs is About $40 Million.

3.1 COAL QUALITY IMPROVEMENT

Issue

The pyritic sulfur and ash content of coal can be reduced substantially, especially for Midwest and Northern Appalachian Coal, through physical cleaning and blending. Today only about one-third of utility steam coal is cleaned to remove some pyritic sulfur. About 80% of this cleaned coal could be cleaned more effectively to increase sulfur removal and improve blending characteristics. The existing EPRI Coal Cleaning Test Facility (CCTF) in Homer City, Pennsylvania, provides the necessary focal point for resolving this limitation.

Response

- Improve conventional physical coal cleaning efficiency and cost by improving dewatering/drying, process control, and performance of cleaning processes (froth flotation, heavy media cyclones, concentrating tables for recovering the fine coal fraction. ($35M, 1984-1987)

- Develop advanced coal cleaning processes capable of cost-effectively producing lower sulfur and ash coal fractions. The focus will be on improved comminution, drying, storage and handling methods including coal slurry production and handling. ($30M, 1984-1988)

- Western low-rank coal beneficiation to reduce alkali metal content. This limits severely the degree of blending with Eastern high sulfur coal that can be achieved because of adverse effects on fouling and slagging. ($15M, 1984-1988)

- Steam coal cleaning and combustion characterization to confidently determine the capability and cost of coal cleaning and verify the operational benefits of quality changes which result. This would involve a total of 50 steam coals. ($25M, 1984-1988)

Funding

EPRI Budget	$ 40M	(1984-1988)
Total Required	$105M	(1984-1988)

Deliverables

Application guidelines and commercially demonstrated process improvements for cost-effectively increasing the breadth and intensity of steam coal cleaning. Techniques for improving the blending potential of low sulfur Western coal with high sulfur Eastern coal.

3.2 FURNACE LIMESTONE INJECTION

Issue

Need to qualify furnace limestone injection, establish data base, range of applicability and site-specific cost factors for commercial retrofit applications.

Response

- Pilot-scale studies (incorporate in prototype tests if promising). Also investigate furnace limestone injection (FLI) combined with other subsystems; e.g., FLI + NH_3 + ESP, FLI + wet EPS, FLI + spray-dry. ($16M, 1984-1988)
- Construct and evaluate three 20-100 MW prototype installations at heavily instrumented coal-fired utility boilers. Tests will include alternative burners, injection methods, and sorbents at each host unit. Resolve resulting particulate control and ash handling issues. Also use to investigate questions arising out of full-scale demonstrations. ($23M, 1985-1988)
- Full-scale (300-700 MW) retrofit demonstration at two or more existing boilers selected for different boiler designs, etc. Verify removal capabilities, reliability, and plant impacts, effect on particulate collection, water loops and solid waste management. ($50M, 1985-1988)

Funding

EPRI Budget	$11M	(1984-1988)
Total Required	$89M	(1984-1988)

Deliverables

Utility proven technology; comprehensive economic evaluation; proven applicability as a function of boiler design, coals, etc.; known impacts on boiler, particulate control, solids disposal, water balance.

3.3 COMBUSTION NO_x

Issue

Lack of demonstrated retrofittable low-NO_x burner modifications for the range of utility boiler designs. Even developmental low-NO_x burners today are limited in potential to about one-third of the pre-NSPS boiler capacity. Also may be required for effective SO_2 removal via Furnace Limestone Injection.

Response

Follow RP1836 and RP2154 prototype low-NO_x burner evaluation program with full-scale retrofit demonstration at three or more representative boilers of different types--e.g., C-E tangential, front-wall fired, B&W cell, etc. ($36M, 1984-1988)

Funding

EPRI Budget	$ 7M	(1984-1987)
Total Required	$36M	(1984-1988)

Deliverables

Design and operating guidelines, cost evaluation, proven range of applicability. Commercial availability from several suppliers. Increase practicality of furnace limestone injection process.

3.4 RETROFITTABLE PARTIAL SCRUBBING

Issue

Limited information base for retrofit design as well as operating and cost impacts of partial scrubbing (less than 90% removal).

Response

- Prototype (10-20 MW) testing, similar to TVA/EPA/EPRI Shawnee test programs:

  --Simplified contactors; i.e., if high SO_2 removal is not required, can contactors be designed for lower maintenance cost and higher reliability?

  --In-duct spray dry SO_2 removal. ($10M, 1984-1987)

- Updated economic evaluation of partial scrubbing:

  --50-90% SO_2 removal, variable, at a single existing or newly designed scrubber.

  --New scrubber designed to 50% vs. 70% vs. 90% removal. ($5M, 1984-1988)

- Design, construct, operate a full-scale (100-200 MW) demonstration at existing coal-fired plant. Test the concepts tested at prototype scale above, plus others that can only be tested at full-scale, e.g.:

  --Mixed reheat and by-pass strategies.

  --Partial SO_2 removal by coal additives (could include with furnace limestone injection demos. ($45M, 1984-1988)

Funding

EPRI Budget	$ 5M	(1984-1988)
Total Required	$60M	(1984-1988)

Deliverables

Engineering and economic guidelines for applying partial scrubbing as a retrofit emission control option.

3.5 CHIYODA CT-121 FGD RETROFIT DEMONSTRATION

Issue

Chiyoda jet bubbler reactor scrubber offers improved reliability, a dry gypsum by-product, simplified retrofit potential over conventional limestone FGD. Prototype tests completed but not yet commercially demonstrated for utility application.

- 23-MW prototype tests successful on medium sulfur coal (EPRI/Southern Company)
- Commercial retrofit has been installed at a heavy oil-fired petrochemical plant in Japan (Mitsubishi Petrochemical)
- 60-MW retrofit demonstration is planned on a high sulfur, coal-fired boiler (University of Illinois)

Response

Full-scale (150-200 MW) retrofit installation and test program at a utility site using high-sulfur and high Cl/F coal. Design for continuous 90% removal (NSPS) with closed water loop (zero-discharge). ($35M, 1984-1987)

Funding

EPRI Budget	$0.5M	(1984-1985)
Total Required	$ 35M	(1984-1987)

Deliverables

Commercially demonstrated, reliable, compact retrofittable FGD system, documented chemistry and materials behavior, operating costs.

3.6 ADVANCED PROCESS EVALUATIONS

Issue

Advanced SO_2 and SO_2/NO_x removal techniques that now appear marginally attractive for 90% SO_2 removal at new plants due to high cost, could be relatively more attractive at lower removals due to retrofittability or different cost-vs-removal sensitivity.

As the demand for retrofittable control options increases, some of the less-developed processes may be pushed on the political front without an adequate technical and economic information base to back them up.

Response

- Pilot (1-2.5 MW) evaluation testing on coal-fired flue gas (similar to spray-dry FGD and SCR de-NO_x projects at Arapahoe) to establish operating sensitivities, performance capabilities, and identify plant impacts. Follow-up with prototype (10-20 MW) verification where warranted. ($20M, 1984-1988)

- Engineering and economic evaluation per EPRI premises, of processes selected for pilot testing. To be done in parallel or overlapping; i.e., begin economics as soon as data start to become available. Level II analysis for those selected for prototype verification. ($5M, 1984-1988)

Example Processes

a. Limestone dual alkali (FMC)

   --Retrofit to existing limestone FGD.

   --Retrofit to existing lime dual alkali (limited applications).

b. Activated carbon/NH_3/coal reduction SO_2/NO_x (Mitsui, Bergbau-Forschung, Sumitomo/IHI)

   --Appears technically feasible but not economic for high-sulfur coal, high SO_2 removal; economic trade-offs need exploring.

c. Sulf-X, Avco/Ebara

--Appear technically sound, could be attractive retrofits if costs could be cut.

d. Bischoff

--Gaining momentum in Germany with industrial applications.

Funding

EPRI Budget	$ 4M	(1984-1988)
Total Required	$25M	(1984-1988)

Deliverables

Guidelines for selection of processes, and economic evaluation for retrofit at 50-90% SO_2 removal.

Credible data base for design specifications, focusing on retrofit applicability at lower removals.

3.7 WATER & SOLIDS INTEGRATION

Issue

Little attention has been given to date to secondary pollution questions of acid rain mitigation, notably compliance with Clean Water Act/NPDES and RCRA. In addition to a very large increase in both solid and liquid waste products created by retrofit control requirements, scrubber chemistry will be different at lower removals. New approaches will produce by-products of different characteristics--e.g., more soluble calcium in waste solids from furnace limestone injection than conventional wet limestone scrubbing.

Response

In conjunction with planned EPRI programs for integration of the cooling tower with the SO_2 scrubber loop plus FGD chemistry, sludge disposal and groundwater protection, this project would construct, operate and evaluate a full-scale integrated water treatment and solids management system at one or more existing plants. The system will focus on retrofit applications and be designed along with retrofit SO_2 controls. (This work extends the scope of EPRI programs now planned under RP1031, 1261, 1406, 1646, 2114.) ($45M, 1985-1988)

Funding

EPRI Budget	$ 8M	(1984-1988)
Total Required	$45M	(1984-1988)

Deliverables

Demonstrated application of retrofittable water quality control technology. Design and operating guidelines for retrofit water and solids systems.

3.8 ELECTROSTATIC PRECIPITATOR PERFORMANCE IMPROVEMENT

Issue

The retrofit application of in-furnace or certain other flue gas emission control technologies will have a detrimental effect on the performance and reliability of existing electrostatic precipitator (ESP) facilities at these plants. This degradation will result from both the increased particulate loading as well as the effects on fly ash resistivity, adhesivity, and particle size resulting from changes to fly ash and flue gas composition by the retrofitted control processes.

Response

Upgrade existing ESP technology including development of improved flue gas conditioning additives, pulsed power supplies, on-line performance diagnostic instrumentation and improved fly ash properties prediction methods. ($9M, 1984-1986)

Full-scale field evaluation of retrofit effects plus verification of performance improvements. ($16M, 1985-1988)

Funding

EPRI Budget	$ 5M	(1984-1988)
Total Required	$25M	(1984-1988)

Deliverables

Utility proven techniques for maintaining and upgrading ESP performance and reliability. Guidelines for the operation and maintenance of the upgraded ESP facilities.

3.9 FLUIDIZED BED CONVERSION

Issue

Fluidized-Bed Combustion (FBC) has developed to the point where it can be a viable retrofit utility option to reduce SO_x and NO_x emissions as well as reduce sensitivity to coal quality. Of particular interest is the potential for conversion of up to 45,000 MW (e) of existing, pre-NSPS pulverized coal furnaces to FBC. The confidence to achieve this potential for improved environmental performance and fuel flexibility will depend on successful demonstration of such a conversion.

Response

Implement utility proposal to convert an existing 100 MW (e) boiler to AFBC and demonstrate its reliability and economically competitive performance. This demonstration will be based on the extensive pilot and prototype FBC development efforts conducted by EPRI over the past seven years.

Funding

EPRI Budget	$15M	(1984-1988)
Total Required	$60M	(1984-1988)

Deliverables

Demonstrated ability to convert a utility boiler to FBC and operate it in compliance with both environmental and economic performance requirements.

Section 4

OPTIONS FOR THE 1990s

At the end of this decade we would expect that scientific research will be continuing on the environmental effects of power plants. Many of today's questions will have been answered but new ones will arise. A number of new coal based options should however be available for the generation of electricity. EPRI has been supporting the development of coal gasification combined cycle systems and the atmospheric fluidized bed combustion. These options should be available by the end of the decade. Three of the EPRI programs are specifically directed to the development of these options:

- Clean Gaseous Fuels
- Fluidized-Bed Combustion and Alternate Fuels
- Long-Range Alternatives

4.1 CLEAN GASEOUS FUELS

Program Description

The goal of the Clean Gaseous Fuels Program is to develop economically competitive and environmentally acceptable coal-gasification-based generating systems.

Intermediate Btu gas can be fired directly in boilers or, taking advantage of its cleanliness, in combined cycles, where the turbine or fuel cell exhaust is used to raise steam for a steam turbine/generator. Integrated gasification combined-cycles (IGCCs) have significant potential as baseload, coal-fueled power plants that can meet stringent environmental standards.

EPRI studies indicate that integrated gasification-combined cycles using commercially available combustion turbines (2000°F inlet temperature) and based on Texaco, Shell or BGC/Lurgi gasifiers are potentially competitive with conventional coal-fired power plants with stack gas cleanup.

The integrated gasification combined-cycle power plant promises reduced heat rates, lower water consumption and land requirements, markedly reduced emissions, and capital and electric power costs competitive with conventional coal-fired steam plants.

The principal project in the Clean Gaseous Fuels Program and the major focus for all development efforts is the Cool Water GCC demonstration. EPRI initiated this project with Texaco and Southern California Edison in late 1977. The objective is to design, build and operate an integrated gasification combined-cycle demonstration plant of about 100 MW by 1985. The program also includes work to improve gasifiers, gas cleanup technology, heat recovery boilers, and other components of gasification-based generating systems.

Funding

EPRI Budget ____$76.6M____ (1984-1988)

Deliverables

Demonstrated coal gasification combined cycle systems for application in the latter part of this decade.

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4.2 FLUIDIZED COMBUSTION AND ALTERNATE FUELS

Program Description

A primary goal of the Fluidized Combustion and Alternate Fuels Program is to develop fluidized-bed combustion boilers that will reduce the cost of an environmentally acceptable electricity and allow utilities to use lower grade fuels.

A second goal is the development of a coal slurry fuel and firing system that will permit clean coal to be substituted for oil in some oil-designed boilers.

This program focuses on developing the information needed to build a large-scale (about 200 MW (e)) AFBC boiler and its auxiliaries, ready to generate steam for a new or existing turbine generator on the TVA system by 1989.

Detailed planning for the 100-200 MW (e) demonstration project started in 1982 with the selection of several plant sites, specific applications and cosponsoring utilities to develop detailed project plans. To maximize project viability, the demonstration AFBC could be designed to: (1) burn municipal solid waste, bark, or any other combination of low-grade fuels, (2) repower an operational oil-fired plant, and (3) add an increment of steam supply to compensate for a poorly performing pulverized coal boiler, possibly with the addition of a topping turbine.

Studies have also been underway on the development of a turbo-charged boiler and ultimately a coal-fired combined-cycle system, based on pressurized fluidized-bed combustion (PFBC). A PFBC turbocharged boiler may offer major capital costs and environmental advantages, vis a vis conventional PC/FGD plants. Operation at lower gas temperatures (800°F) minimizes the technical problems limiting immediate commercial deployment, specifically development of a compatible dust removal/gas turbine system that will operate reliably.

Funding

EPRI Budget $111.8 M (1984-1988)

Deliverables

Demonstrated Atmospheric Fluidized Bed technology for commercial application in the latter part of the 1980s.

4.3 LONG-RANGE ALTERNATIVES

Since this plan addressed a period of forty to fifty years, all of the potential contributors to the reduction of sulfates and nitrates in the atmosphere must be recognized. Examples of alternatives that could reduce emissions would include a revitalized nuclear power option, renewable resources, and fuel cell technology.

EPRI research and development on the nuclear option is primarily directed towards improving the reliability of existing plants, assuring the safety and demonstrating the economics of this mode of generating electricity. The experience gained from the first generation of the light water reactors could be used to design and build more reliable and less costly systems. The R&D program now underway addresses these subjects in a systematic way so that confidence could be rekindled in this technology. International progress in building and operating nuclear units is also being examined to establish the lessons to be learned from the experience of other nations.

The use of fuel cells could result in essentially no emission of SO_2 and NO_x. EPRI supported R&D on the United Technology Corporation (UTC) phosphoric fuel cell technology is in the prototype testing stage. Startup tests are underway on a 4.8 MW fuel cell unit on the Consolidated Edison system in New York City. A UTC fuel cell system based on essentially the same technology has recently begun operation on the Tokyo Electric System. The economics and the reliability of this technology should be established within the next several years. The application of the fuel cell introduces the question of the availability of fuel. If natural gas proves to be an available and cost-effective source of energy for the next few decades, fuel cells will be applied using the energy source. Other fuels including coal gasification are being studied for this application.

EPRI R&D addresses a broad range of technologies based on renewable resources. Solar thermal and photovoltaic systems are examples of subjects that have been supported. Advanced photovoltaic systems will receive research support over the next five years. Wind is currently receiving significant attention by U.S. utilities and by EPRI. Some 930 solar and wind electricity generation experiments are currently underway in the utility industry. These sources of energy must be considered as a limited but important possibility for the future.

EPRI research and development on storage systems could also lead toward economical methods of increasing the capacity factor of the newer baseload plants. Such units will typically emit fewer precursors of acidic deposition than the older coal-fired ones. Storage systems would reduce the amount of coal and oil required for the peak period. Compressed air, hydroelectric and battery systems are being studied. Substantial laboratory and test work is underway on battery systems that should be of value in specific locations in the country.

These technologies together with the advanced coal-based alternatives offer a future in which atmospheric sulfates and nitrates resulting from utility system operations could be substantially reduced. In all cases, the economics of the reduction must be considered so that a balance is struck between the value of the added improvements and the cost of adoption of each new alternative.

STATEMENT
David R. Williams
Chairman of the Board
Integrated Carbons Corp
to the
House Science and Technology Committee
Subcommittee on Energy Development and Applications,
and Natural Resources, Agriculture Research and Environment
September 20, 1983

I am submitting the attached statement pertaining to Purged Carbons™ for consideration by you and your Subcommittee in connection with your hearing on Acid Rain. The subject of the hearing is of great interest to us, and we felt that your Subcommittee would be interested in the Purged Carbons™ technology, which has the potential for providing an alternative to foreign oil and other fuels using domestic coal in a clean - burning coal - based fuel that does not require scrubbers and expensive retrofitting and is favorably competitive with the current price of oil.

There currently is great international interest in acid rain. The ultimate objective of Purged Carbons™ technology is to produce an economically competitive fuel so clean that it will meet air quality requirements without scrubbers or precipitators. This goal requires organic sulfur removal. Integrated Carbons Corporation is contracting to develop a microbiological process for oxidizing the resistant cyclic thiophenes in coal. The process as presently configured goes a long way toward reducing particulates and SO2. Slurry firing with its lower combustion temperature will emit far less NOx. Information on NOx reductions by slurry firing comprises a specific goal in the test program.

American industry has a large inventory of gas and oil-fired boilers, heaters, dryers, and kilns. The cost of replacing these units with new coal-fired equipment is not attractive. For this reason an ultra clean coal, which utilizes the existing liquid transportation and storage systems and can be burned cleanly in these units is highly desirable.

As the price of oil increases and its supply and reserves decline, a shift to coal-based energy will be increasingly desirable for all power plants, including gas turbines and diesels.

Current coal utilization technology is not acceptable in today's industrial applications and urban utility plants of tomorrow. Significant improvement is required in coal handling, storage, sampling, and burning to minimize problems of air quality, spontaneous combustion, unit availability and maintenance costs. The most logical answer to the above problems is Purged Carbons™ fuel delivered as a coal water mixture.

Among other uses, Purged Carbons™ can displace #6 fuel oil in utility and industrial boilers. The substantial potential for this use is apparent. Purged Carbons™ is potentially valuable in national defense as a military fuel, where its benefits include domestic availability, and when used in military vehicles, a high resistance to explosion upon impacts. The process also has potential for producing other non-fuel by-products, such as activated carbons, carbon black, and possibly graphite, among others.

Another major benefit of the Purged Carbons™ process is that it can be applied to all U.S. coal supplies, whether high sulfur or low sulfur, Eastern or Western, bituminous of subbituminous. Processing plants can readily be developed in any of the Nation's coal-producing areas, allowing for dispersal of facilities and the attendant economic and employment (as well as defense) benefits. The process may aid in the development of coal supplies previously considered too remote or uneconomic to produce.

Integrated Carbons Corporation expects that with sufficient developmental support it can produce Purged Carbons™ on a commercial scale within this decade, beginning at a rate approximately equivalent to 60,000 barrels of oil a day and escalating rapidly from there, depending on the number of plants built.

We believe that environmental impacts of the production process can be kept to a minimum. Acids will be produced on-site and will be recycled, so there will be no problems of transportation of acids to and from the plants. There will be no liquid waste stream from the Purged Carbons™ itself, combustion gases, and ash residur. The basic technology is known and can be controlled. During the second stage of the development program, upon which we are now embarking, we will be monitoring very carefully all aspects of the operation, emissions and products, to assure a safe process and cost competitive product once commercial scale is attained.

Upon completion of the development process, we fully expect that we can supply commercially a coal-based oil substitute for a cost in the range of $3 to $4 per million Btu, below even the current cost of oil. The cost advantage should increase with time.

Recently, the Integraged Carbons Corporation completed Stage I of the Purged Carbons™ development program at a cost of over $2 million, borne entirely by the Company. Stage I focused on batch process development, patent work, and economics. Stage II of the developmental process is just now underway, and assuming the availability of adequate and timely funding, will focus on the continuous operation of the process, further refinement of economics, and further data gathering with respect to such issues as materials handling, transportation, potential product uses, and environmental impacts. Stage II is expected to cost over $7 million and take 18 months to complete.

The Congress has recognized the strategic need for alternative fuels and has allocated billions for its development. We have a technology which we think might be the answer but requires capital, which is not available today in sufficient amounts from the private sector, to complete its development. The U.S. Synthetic Fuels Corporation has advised, without addressing the viability of the technology, that the technology does not meet the maturity requirements of their program. While we continue to fund our process development with our own resources, we would welcome any suggestions from the

Committee as to how our technology might be evaluated in the context of its potential to our country and appropriate assistance obtained.

Hazen Research, Inc. of Golden, Colorado, the largest independent hydrometallurgical research firm in this country has evaluated the Purged Carbons™ process as having the capability of producing a coal-based fuel which can be directly substituted for foreign oil and other fuels in many applications; which can be produced for substantially less than the current price of oil; and which could be commercialized within this decade.

We are aware that many alternative energy approaches exist, but few offer the potential of the Purged Carbons™ product for making use of the Nation's vast domestic coal resources in meeting its future energy needs, while reducing its dependence on foreign oil and achieving air quality standards. We are excited by the possibility of assisting the Nation in achieving the important goals of the Energy Security Act of 1980 (PL-96-294), to achieve energy security for the United States for the health of the national economy, the well-being of our citizens, and the maintenance of national security, in a manner consistent with protection of the environment.

integrated

PURGED CARBONS PROCESS

EXECUTIVE SUMMARY

SEPTEMBER 12, 1983

320 South Boston
Suite 831
Tulsa, Oklahoma 74103
(918) 582-5811

INTRODUCTION

The purpose of this summary is to introduce our continuing program to develop an economically and environmentally attractive coal-based alternative to fluid petroleum fuels which we believe can be on the market within six years.

Accordingly, outlined herein are the technologies, economics, and means of implementing a project to produce a COAL-BASED SUBSTITUTE FOR OIL AND GAS.

One can recognize the potential of this technology by the disparity between the price of crude oil and raw coal, roughly four to one on a thermal equivalent basis. This disparity is even more remarkable when considered in light of the inevitable world energy transition from oil to coal.

We discuss in the following pages the factors that impede this transition, and why our process addresses those impediments and takes advantage of this disparity. We describe a new clean coal product, PURGED CARBONS™ which will allow coal to enter the higher end of the fuels market heretofore available only to oil and gas. While it is now viewed as an oil and gas substitute, primarily; its cleanliness and relative low processing cost may make it attractive as an alternative to coal precluding expensive plant modifications necessary to limit SO_2 and NO_x emissions and thereby Acid Rain. Regardless of its range of uses, Purged Carbons™ will create incremental damand for coal by expanding its use to higher grade markets. The process has the potential of making the transition away from overdependence upon petroleum and providing national self-sufficiency in energy supply.

RATIONALE

Coal is abundant and sells at one-fourth the price of oil. Yet 80% of the energy consumed in the United States is petroleum-based. The central reason for this seeming contradiction is that we built our energy infrastructure around the use of oil at a time when it, too, was inexpensive. As a result our environmental regulations as well as our combustion systems are based on oil specifications.

Coal, relative to oil, is a dirty fuel. It is a solid while oil is a liquid. Therefore, to use coal in place of oil, coal must be cleaned to meet oil standards and presented as a fluid to oil-designed combustion systems. Both high purity and fluidity must be achieved at a price which is competitive with oil.

There are three basic approaches to creating a coal-based oil substitute; direct conversion to a liquid (synthetic fuel), physical cleaning, and chemical cleaning.

Direct conversion of coal to an oil-like liquid requires the addition of hydrogen to the coal which is produced by the reduction of water. The oxidation of a substantial portion of the available hydrocarbons in process is a penalty of direct conversion. Direct conversion processes, while some produce a clean liquid fuel, have consistently failed to qualify as economically viable alternatives to oil due to low conversion ratios as well as high capital costs associated with the high pressure and high temperature systems they employ. Historically the price of coal has risen to reflect increases in the price of oil. This sympathetic movement has consistently proven all conversion technologies uneconomic regardless of the extent of oil price increases.

Physical cleaning of coal depends upon the breaking away of mineral matter from the hydrocarbons in coal. Because coal is formed by the random sedimentation of hydrocarbons and minerals, maximum liberation requires reducing the particles to a very small size. On a batch basis, mineralization has been reduced to slightly less than 1% of the total coal weight by one physical cleaning process under development. While estimated costs allow production competitive with oil, the 1% ash levels attained are far from oil standards. This process requires the use of freon and the micropulverization of the coal, which present environmental and handling challenges which must be overcome prior to commercialization.

In the area of chemical cleaning, the only reagents known that effectively separate ash from coal at ambient temperature and pressure are a combination of hydrofluoric and hydrochloric acid. Mineral matter may be quantitatively determined by HF and HCl dissolution, according to a U.S. Bureau of Mines procedure developed in 1959.1/ No other chemicals dissolve the ash-forming minerals without negatively affecting coal's combustion characteristics.

(Processes which might be considered competitors to Purged Carbons™ are discussed in Appendix C; they include molten alkali and oil (or freon) agglomeration processes.)

Although HF/HCl treatment is generally known to nearly eliminate ash, since the Second World War, when the Germans used HF and HCl as a precursor to synthetic fuel production, serious attempts to commercialize an acid-based beneficiation program have been limited. Low oil prices discouraged interest in alternative fuels until the early 1970's. Thereafter, most attention has been focused on direct conversion technologies in anticipation that rapidly rising oil prices would support the very high costs associated with these processes. Lack of attention to the potential of HF/HCl cleaning is understandable as no coherent scheme addressing economic and environmental issues for the use of chemically cleaned coal has, until now, been developed.

Integrated Carbons Corporation was formed specifically to explore the potential of chemically reducing the ash and sulfur levels of coal sufficiently to make it economically and environmentally competitive with oil. Its Purged Carbons™ process, which employs HF and HCl leaching, is now sufficiently mature to present as the keystone of a comprehensive mine-to-boiler system, which can seriously be considered as a viable means of replacing oil-based products in a range of applications.

WHAT IS PURGED CARBONS™?

Definition

Purged Carbons™ is the hydrocarbon material remaining after the mineral matter (ash) has been removed from the coal by a combined dissolution and gravity process. Typical analyses of Purged Carbons™ are:

1/ Evaluation of an Acid-Extraction Method for Determining Mineral Matter in American Coals", U.S. Bureau of Mines, Report of Investigation 5470, 1959.

Feedstock	Ash %	Sulfur %	Calorific Value Btu/lbs
Eastern Appalachian Coal	0.25	0.80	15,300
Western Subbituminous Coal	0.25	0.25	12,800

Purged Carbons™ can be formulated as a coal-water mixture (CWM) containing 70% solids. This high density, ultra clean slurry can be pumped, piped, metered, and fired like any other high viscosity hydrocarbon fuel, and depending upon feedstock selection it is cleaner than many low-sulfur oils.

Flowsheet

The process for producing Purged Carbons™ employs aqueous hydrofluoric (HF) and hydrochloric (HCl) acids at ambient pressures and temperatures for beneficiation. Ash is the only waste product from the process; it is produced in a pyrohydrolyzer used to regenerate the acids. The Purged Carbons™ product from the process has only small quantities of halogens, comparable to the halogen content of the feed coal. The process flowsheet is given in Figure 1. Process chemistry is reviewed in Appendix A.

UTILIZATION

Purged Carbons™, either dry or slurried, is intended to substitute for gas and petroleum in these areas:

- Boilers designed for #6 oil
- Boilers designed for coal
- Gas turbines
- Diesels
- Non-fuel applications (carbon black, activated carbon, electrode carbon etc.)

To achieve this, there are several challenges which must be addressed in both the short- and long-term:

Environmental restraints demand near zero emissions from power plants. This requires either pre-cleaning of the coal and/or a flue gas cleanup system. Most economic studies support pre-cleaning in lieu of flue gas cleanup systems after combustion.

FLOWSHEET FOR
PURGED CARBONS PROCESS
LOW ASH EASTERN COAL

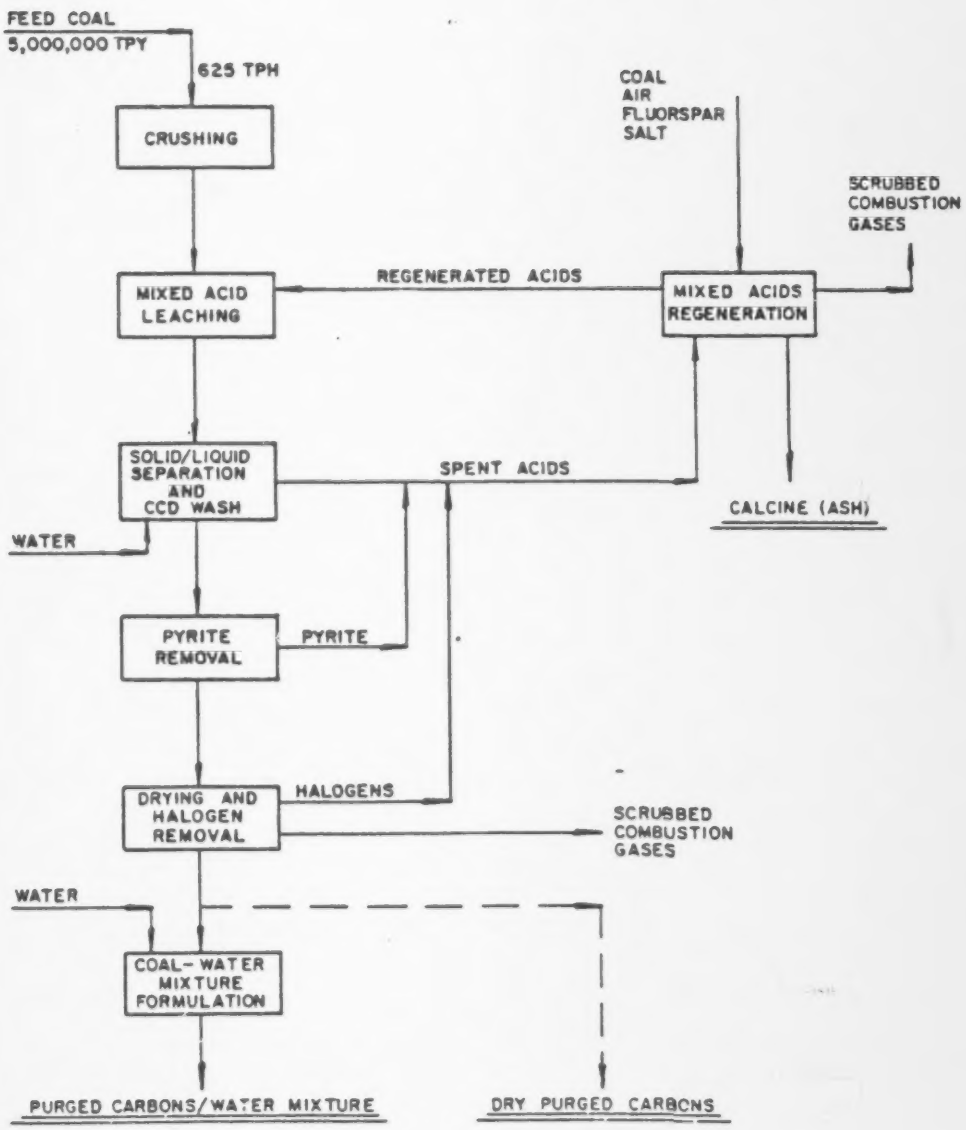

FIGURE 1

There currently is great international interest in acid rain. The ultimate objective of Purged Carbons technology is to produce an economically competitive fuel so clean that it will meet the air quality requirements set forth in the NSPS without scrubbers or precipitators. This goal requires organic sulfur removal. Integrated Carbons Corporation is contracting to develop a microbiological process for oxidizing the resistant cyclic thiophenes in coal. The work will identify suitable organisms and optimize the extraction process. The process as presently configured goes a long way toward reducing particulates and SO_2. Slurry firing with its lower combustion temperature will emit far less NO_x. Information on NO_x reductions by slurry firing comprises a specific goal in the test program.

American industry has a large inventory of gas and oil-fired boilers, heaters, dryers, and kilns. The cost of replacing these units with new coal-fired equipment is not attractive. For this reason an ultra clean coal, which utilizes the existing liquid transportation and storage systems and can be burned cleanly in these units and is highly desirable.

As the price of oil increases and its supply and reserves decline, a shift to coal-based energy will be inevitable for all power plants, including gas turbines and diesels.

Current coal utilization technology is not acceptable in today's industrial applications and urban utility plants of tomorrow. Significant improvement is required in coal handling, storage, sampling, and burning to minimize problems of air quality, spontaneous combustion, unit availability and maintenance costs. The most logical answer to the above problems is Purged Carbons™ fuel delivered as a coal water mixture.

(The rationale for converting from an oil-based economy to a coal-based economy is further detailed in Appendix B.)

ECONOMICS

While the costs of producing Purged Carbons™ varies depending upon factors such as percentage of moisture and ash content, the need for plant winterization, and the number of Btu's available per ton, recent economic evaluations developed by Hazen Research, Inc. estimate that Purged Carbons™ produced from low-ash Eastern U.S. coals will have a process cost including formulation of a coal water mixture of $1.11 added to a feedstock cost of $39/T delivered total cost (without profit) is $2.53 MM/Btu.

SHAREHOLDERS

Williams Resources, Inc.
Robert R. Horton
Investment Partnership
(comprised of the Misener and Bell family interests and clients and partners of Gray, Siefert & Co.)
Anka and Arnick Holdings Limited
Burlington Northern Inc.
Morgan A. Greenwood
Harbour Investments
Sarah Williams

MANAGEMENT

Senior management, which has demonstrated a strong commitment to the project in terms of both a financial investment as well as active involvement, consists of:

David R. Williams, Jr.	Chairman	October 1981
Morgan A. Greenwood	President	October 1981
Robert R. Horton	Executive V.P.	March 1981
Charles T. Ray	V.P. Finance & Administration	February 1982
Dr. James Kelly Kindig	V.P. Technology	January, 1982

Mr. Williams was a founder and served as Chief Operating Officer of The Williams Companies. He has served as Chairman of Williams Brothers Engineering, Holmes & Narver and The Resource Sciences Corporation. During his tenure, the Resource Sciences Group was cited five consecutive years by the Engineering News Record as being the world's leader in design engineering (by volume). Current directorships include Burlington Northern Inc. and Tne Williams Companies.

Mr. Greenwood, until recently, served as President of The Resource Sciences Group and Senior Vice President of U.S. Filter Corporation. Current directorships include Shaw Industries (Canada) and The University of Tulsa.

Mr. Horton founded and served as Chief Executive Officer of Dialog Corporation. He has served as Executive Vice President of Travel Network Corporation and has acted as a principal of the New York Stock Exchange.

Mr. Ray is both a C.P.A. and Attorney. Recently, he has served as the Senior Vice President Finance and Administration of The Resource Sciences Corporation and as Group Vice President of U.S. Filter Corporation.

28-916 0 - 84 - 77

Dr. Kindig is the holder of a number of patents in the areas of coal beneficiation and metallurgy. He is experienced in project development from activities as an independent consultant and as Vice President Coal Technology at Hazen Research, Inc. of Golden, Colorado.

Integrated Carbons Corporation management augments its staff by the liberal use of contracted professionals. In the technical area, Hazen Research, Inc. has been a major contractor. Patent work is pursued by Sheridan, Ross & McIntosh of Denver, Colorado. Outside legal and tax counsel is Donald B. Atkins of Tulsa, Oklahoma. Integrated Carbon's auditor is Arthur Young & Company.

ENGINEERING AND ENVIRONMENTAL CONSIDERATIONS

The Purged Carbons™ process is a hydrometallurgical process, not a coal conversion process. Consequently, it employs the same equipment used by other hydrometallurgical processes; crushers, tanks, stirrers, pumps, thickeners, filters, dryers, etc. The technology for the use of hydrofluoric acid has been perfected through its widespread use in oil refineries in the production of high octane gas. Even the pyrohydrolizer used for acid regeneration is now used by the steel industry for regenerating pickle liquors (HCl). Further, the availability of equipment permits scale-up in minimum time.

Complete material and energy balances were developed by Hazen Research, Inc. The overall thermal efficiency is 96.4% with 3.6% of the energy being consumed as process heat for pyrohydrolysis and drying. The hydrocarbon portion of the coal is altered only as to increase its purity as it passes through the process.

The principle fuel to the pyrohydrolyzer is coal fines. They are carried to the pyrohydrolyzer with the pregnant liquor (spent acids.) This achieves a dual function of supplying heat to the pyrohydrolyzer and de-sliming the leached coal enabling must faster thickening and filtration.

Raw materials for the process are coal, fluorspar, salt, and water. Acids are generated and regenerated on-site. Discharge products are Purged Carbons™ and ash. All gaseous effluents from the plant are scrubbed with a lime slurry which captures all fluorides, chlorides and sulfur dioxide. A bleed stream of the lime slurry is sent to the pyrohydrolyzer for HF and HCl recovery, and for discharge of sulfur as sulfates. There are no liquid effluents scrubbed. Water used to wash the product exits the plant as clean steam.

PROCESS HISTORY

As mentioned earlier, removal of ash minerals from coal by halogen acids has been reported in the literature at various times since the 1920's. In recent years the use of gaseous HF under pressure in a batch mode was proposed by Kinneret[2/], an Australian group.

Beginning in 1982, Integrated Carbons began an intensive process development program in the HF and HCl deep cleaning of coal, as a departure from earlier work done by others. The process was made continuous and atmospheric, and aqueous acids were used instead of acid gas. Economics and process development have both undergone three iterations. Acid recovery evolved through calcium chemistry, to alumina chemistry, to the currently used pyrohydrolysis reactions.

The process and economics for Purged Carbons™ were developed by independent researchers and consultants under the guidance and at the expense of Integrated Carbons Corporation. At the present stage of development, a ±30 percent level of certainty was ascribed to the process economics by Hazen Research, Inc., who developed them.

DEVELOPMENT STRATEGY

Objective

To complete the development of Purged Carbons™ technology while assuring continuity of ongoing responsibility for the enhancement of the technology, Integrated Carbons Corporation plans to be the research and development organization which licences the commercial applications of the technology, provides technology transfer, maintains continuing research and improvements, and provides technical support services to licensee.

Development Program

As mentioned, Phase I consisting of development of the process, lab testing, a batch mode small pilot plant, and a preliminary feasibility study has now been completed, all funded by the existing shareholders.

Approximately $56MM will be required to prove and refine the technology, including non-site specific plans and specifications for a 5MM/TPY plant. The steps to attain this objective are:

2/ Kinneret Enterprises Ltd., Robert Lloyd and Maxwell Turner, European Patent Application 0 016 624, filed March 14, 1980, "Coal De-Ashing Process".

Phase II

A 250 lb/ph continuous pilot plant program. This program will take one year and six million dollars to complete. It will reduce cost uncertainty from the current ± 30% to ± 20%, produce enough product to establish combustion data based upon laboratory and small scale burns, develop slurry, storage, and transportation data necessary to freeze a design and build a 40,000/TPY demonstration plant. The cost of this phase is estimated to be approximately $6 million.

Phase III

The demonstration plant program is estimated to require 18 months and 50 million dollars. At its conclusion, commercial test burns of Purged Carbons™ based products, complete engineering for a commercial plant, mine to combustion logistical planning, and definitive cost figures will be in place. Additionally, work on non-fuels applications will be initiated with the cooperation of companies in the businesses for which Purged Carbons™ might prove a more economical feedstock. A schematic design for a full commercial plant, on a non-site specific basis, will be achieved as part of this phase.

Whereas the Purged Carbons™ process, as is, represents a viable means to replace petroleum with coal on a cost effective basis, ongoing efforts are being made to further reduce ash and sulfur in the end product without adding substantial process costs. Several promising approaches are in hand and are currently being evaluated. Funding for concerted pursuit of these potentials will be raised for the pilot and demonstration plant programs, Phases II and III above.

FUNDING STRATEGY

Sale of U.S. license: As Integrated Carbons does not intend to build an operating organization, the sale of licenses for various regions of the world will be the main source of future income to the company after completion of the technology from the present stage through the above Development Program.

Funding for Phase II above will be the subject of an R&D Limited Partnership now under preparation, with Integrated Carbons acting as the General Partner. Funding for Phase III above is expectd to be provided from advance royalties from licnsees for the process selected for various geographic regions of the world. Licensees would pay certain advance royalties that would be modest at first until certain levels of confidence were reached during the progress of Phases II and III above.

If performance and cost projections are met, the licensees would pay additional advance royalties if they wished to proceed. Thereafter, the licensee would have access to all improvements, and pay a royalty to Integrated Carbons for all finished Purged Carbons™ produced.

In the orderly pursuit of commercialization, Integrated Carbons has staged the work both in terms of the scale of operation and level of economic certainty; the stages are included in Table 1, Appendix D.

The first stage of development (Bench - Stage I) was completed in June, 1983 at a cost to Integrated Carbons of two million dollars in cash as well as the uncompensated executive time of all but technical staff. The first stage has been evaluated as sufficiently promising to warrant Integrated Carbons' embarking upon the second stage of development (Pilot - Stage II) involving continuous operation, which is hoped will increase the economic predictability from a ± 30% to a ± 20% variability level.

THE PILOT PLANT DEVELOPMENT - PHASE II

Achieving A Level of Certainty to ± 20%

The information which is essential and must be acquired in order to complete the economics to the ± 20% level is process data derived from a continuous plant operation. To that end, a small pilot plant will be built and operated and the required data gathered, thereby enabling the economic study to be completed.

In addition, the transportation and utilization of Purged Carbons™ will be assessed at the same time that the process data are being secured. Purged Carbons™ will be produced on a batch basis at the outset of this development stage (Pilot - Stage II) and dispensed to parties in both the public and private sectors who have already expressed interest in its performance in boilers, turbines and diesels. The early assessment of Purged Carbons™ product is critical because Purged Carbons™ is a totally unique product.

The patent strategy, evolved during the first stage of development, will be further maturated by a combination of experimental and legal work. (Seven patent applications have been filed to date.) The company's patent counsel is Sheridan, Ross & McIntosh of Denver, Colorado.

The impact of Purged Carbons™ production, transportation and utilization on the environment, will be investigated so that appropriate tests can be executed to provide the information required. Additionally, all aspects of permitting will be described to enable merging technical and permitting activities without delays.

One year will be required to complete all work needed to achieve a ± 20% level and meet the objectives of this stage of development; additional data descriptions regarding the above follow.

Continuous Three Ton/Day Pilot Plant

The objectives of the pilot program are to:

1. Prove the technical feasibility of the process on a continuous basis including the reprocessing of recycle streams.

2. Provide engineering data, such as mass flows, analyses of plant streams, solid/liquid separation rates, corrosion and erosion data, etc.

3. Produce material for characterization including chemical analyses and other indicated testing. Of concern here are all plant discharge products; in addition to Purged Carbons™, they are alumina, anhydrite, mixed oxides (ash), silica and neutralized sludge.

The pilot plant will have a feed rate of three tons per day which is 250 lbs/hr. The pilot plant is planned to be disengaged at selected points for convenience. Comminution and beneficiation of the feed coal will take place at a time and rate independent of the leaching circuits. The resulting cleaned and sized coal will be stored in a bin until a later time when it will be sent to the leaching circuits, probably at a different rate. HF leaching, HCl leaching, washing, gravity separation and, if required, baking will be carried on simultaneously. Pregnant liquors from the leaching circuits will be separately stored, and the HF and HCl regeneration circuits will utilize these feed liquors probably in an open circuit that does not return the HF and HCl to their respective leaches.

Operation of the pilot plant will be by campaigns. Operation will be on a 24-hour basis until some problem suggests a shut-down. During shut-down, operation will be on an 8-hour day. Plant streams and products will be analyzed, the problem isolated and fixed, and a new campaign initiated on a 24-hour basis. This mode of operation will continue until the plant can be run successfully, producing specification products for an extended period.[3/]

3/ The number of days will be specified after the plant residence time is established. Generally, pilot plants operate for between 50 and 100 cycles.

Comprehensive chemical analyses will be made on all in-plant flows, mass balances will be secured, and energy requirements estimated. All slurries requiring solid/liquid separation will be bench tested, including where practicable, evaluations with equipment manufacturer's pilot devices. Corrosion/erosion studies will be made with plant materials (solids, liquids, gases and mixtures thereof).

Commercial engineering firms will be canvased for an expression of interest in the design and construction of the 120/TPD (40,000/TPY) demonstration plant (part of the Demonstration, Stage III program). One will be selected from those interested and retained on an advisory capacity to ensure that all data required for the design of a demonstration plant are secured from the pilot plant while it is operating.

Ongoing Laboratory Program

A laboratory program will be maintained throughout the pilot development stage. The principal activities of the laboratory program are:

1. Support for the pilot plant operation. This includes everything from amenability studies of potential feed coals to rationalizing any process chemistry which might prove troublesome.

2. Experimental support for the continuing patent work. The basic patent strategy has been formulated and counsel is proceeding to develop the desired network. Additional tests are desired to further define limits of patentability and distinguish from prior art.

3. Process improvement and innovations. During this development phase, the learning curve is steep; generally not only are process refinements set forth, but also breakthroughs can be made, greatly simplifying or improving a circuit or even the entire process.

Special emphasis is being placed on developing a method for removal of organic sulfur. Methods integral to the process and sequential to the process are both under consideration.

Transportation and Handling

Numerous options among currently practiced methods exist for transporting Purged Carbons™. The selection of the transportation method for a particular case is principally one of economics. They hinge primarily upon:

1. Particle size; three possibilities are:

   a. 1/2-mm by zero, the size produced by the process,
   b. fine powder, less than 200-mesh, the size at which Purged Carbons™ would be used in boilers or heat engines, and

c. pellets, 1/2-inch agglomerates of Purged Carbons™ held together by a binder; pelletized Purged Carbons™ would only be produced if it facilitated transportation in railroad cars or other conveyance.

2. Distance from producer to user or from producer to a delivery point for secondary distribution.

3. The facilities which the user has for storing, handling and using Purged Carbons™; for example, hydraulic pipelining might be preferred if the user were equipped for combusting a slurry.

During this second stage of development (Pilot - Stage II), Integrated Carbons will appraise the relative merits of the numerous transporation methods. The handling, storing and reclaiming of Purged Carbons™ in systems designed for liquids (pipes, pumps, tanks, tankers, etc.) will be the main focus of the work. All systems will be assessed to uncover problems (if any) and provide solutions. The economics for various systems also will be developed.

Development or determination of safe methods for handling and storing Purged Carbons™ is an imperative. The possibility for dust explosions and the potential for spontaneous ignition through self-heating will be analyzed and limits established which will define the areas of operation to preclude them. The use of an oxygen depleted gas (flue gas) for handling Purged Carbons™ dry is the likely method for control. Formulation of Purged Carbons™ in a coal water mixture will also be investigated.

Minus 200-mesh (<74-micro meters) and minus 5-microns (<5-micro meters) probably will be the size of Purged Carbons™ when utilized by industry. Integrated Carbons has made initial estimates of the cost of grinding Purged Carbons™ from minus 1/2-mm (its size when discharged from the production plant) to 80% minus 200-mesh, for power plant use and 80% minus 5-microns. These cost studies will be updated and expanded to include different coals and different size consists as dictated by utilization studies.

Utilization

As a substitute for petroleum products, Purged Carbons™, dry and/or in slurries, will be combusted in:

1. Boilers designed for #6 oil

2. Gas turbines

3. Diesels

Purged Carbons™ also has potential in the non-fuel markets including electrode carbon, carbon black, graphite and activated carbon feedstocks. Further, there is some possibility that Purged Carbons™ may be a better bargain than coal in boilers designed for coal due principally to large reductions in maintenance costs and the cost of replacement electricity (generally produced from oil or gas).

An active program assessing the suitability of Purged Carbons™ to each of the roles sketched above is contemplated during the second (Pilot - Stage II) development stage. Ton samples of Purged Carbons™ will be produced in a batch plant to supply vendors, development laboratories and others interested in the product. These efforts will require a wide range of experimentations, and for some applications Eastern bituminous feedstocks may be better suited while for others, Western subbituminous.

As the program proceeds, Purged Carbons™' samples obtained from the continuous pilot plant will supersede the batch plant samples. Toward the end of this stage of development, utilization tests will be made with Purged Carbons™ produced on a continuous basis.

Economic Evaluation

Project economics are at the core of this development stage (Pilot - Stage II) and the bulk of the program pivots on acquiring whatever data are needed to establish a basis for the economic analyses which will be within ± 20%. Therefore, the personnel responsible for the economic analyses will be intimately involved in planning the pilot plant design and data gathering activities.

In addition to process economics, an outgrowth of the pilot activity, Integrated Carbons will also address the economics of transporting, handling, pulverizing, and utilizing Purged Carbons™.

Project Results

A formal report of the results will be prepared by Integrated Carbons at the end of Stage II of the project. All phases will be integrated into a total assessment of the technology from purchase of the coal through production to utilization of Purged Carbons™.

Proceeding from the second (Pilot - Stage II) to third (Demonstration - Stage III) stage of development will be based upon defined parameters for definition of a viable project -- "technically sound and economically feasible". More specifically, the decision criteria will be resolving of all technical problems and achieving an economic internal rate of return at least 15 percent.

Management, Staff and Facilities

The technical program will be directed by Dr. J. Kelly Kindig. He maintains an office in Denver, Colorado and will have a staff of two professionals and one office person in addition to contracted professionals.

The pilot plant will be built and operated by Hazen Research, Inc. of Golden, Colorado. Hazen Research did the technical work for the Batch - Stage I development. Employing more than 100, it is the largest independent hydrometallurgical research and development firm in the U.S.

As previously mentioned, a commercial engineering firm will be retained in an advisory role to ensure that all data required for design, construction, and operation of the demonstration plant are secured by Hazen. Numerous additional consultants will be utilized in conducting the work, especially in these areas:

1. Commercial application of HF and HCl chemistry.

2. Transportation and handling of Purged Carbons™.

3. Utilization in:

   a. Boilers
   b. Heat engines
   c. Non-fuel uses

4. Environmental and permitting requirements.

5. Selected technical areas, e.g. thermodynamics, materials of construction, alumina industry, etc.

The efficacy of substituting Purged Carbons™ for oil in boilers designed for oil will be assessed in laboratories and bench scale test boilers owned by one or more of the five major U.S. companies who manufacture the combustion equipment used by utilities and industry. Demonstrating the compatibility of Purged Carbons™ with boilers designed for oil is a priority item and all categories of available resources will be mustered to achieve this goal up to the limit of Purged Carbons™ available for test burns.

It is contemplated additional R&D work will be done at other laboratories such as Southwestern Research, Inc., Battelle Memorial Institute, Colorado School of Mines and others for special analysis and confirmation of results. Also, Integrated Carbons will foster arrangements with selected vendors providing incentives for their developing equipment utilizing Purged Carbons™. Government laboratories will be consulted for their interest in fostering Purged Carbons™ utilization.

Appendix A

PROCESS CHEMISTRY

- Dissolution of the minerals associated with coal can be represented by reactions between the oxides and hydrofluoric acid, reading from left to right, as follows:

$$6HF + SiO_2 \longrightarrow H_2SiF_6 + 2H_2O$$

$$6HF + Al_2O_3 \longrightarrow 2AlF_3 + 3H_2O$$

etc. etc. etc. etc.

Regeneration of acid in the pyrohydrolyzer involves forcing these reactions from right to left by the application of heat in the presence of water.

Additionally, sulfur dioxide is produced in the pyrohydrolyzer by burning pyrite. It and oxygen react with calcium fluoride or sodium chloride to produce make-up acids as follows:

$$CaF_2 + H_2O + 1/2\ O_2 \text{ -----> } CaSO_4 + 2HF$$

$$2NaCl + H_2O + SO_2 + 1/2\ O_2 \text{ -----> } Na_2SO_4 + 2HCl$$

The only fluoride lost from the system is that exiting the pyrohydrolyzer in the ash as MgF_2 and the small amount adhearing to the coal.

Appendix B

RATIONALE

The following informational items relate to the inevitability of a world energy transition from oil to a coal-based fuel akin to Purged Carbons™.

1. In spite of an "oil glut", the world has been using more petroleum annually than it has been finding since 1980. Therefore, the end of petroleum use at current quantities and price can be projected as a certainty.

2. The industrialized free world is a huge net importer of petroleum energy from unstable and adverse sources. (70-80% of all Soviet export revenue is from energy sales).

3. The only politically acceptable and sufficiently abundant alternative to petroleum in the intermediate term is clean coal.

4. Environmental activism is not a fad; it is a trend worldwide. Coal, if it is to be burned in the future, must be cleaned to meet oil emission standards. It also must be cleaned in a way that produces only environmentally acceptable waste during its production. (For most applications, this means by processes imposed prior to combustion.)

5. In order to receive market acceptance, any clean coal product must be economically competitive with oil.

6. Commercial energy users in the United States should be motivated to use clean coal products as the depletion of petroleum does not make it eligible as a long-term supply base, and in the interim, petroleum supply can be severely impacted by foreign political events.

7. Clean coal products, with abundant and secure sources of supply, can be contracted on a long-term basis which is not subject to interruption. (90% of all fossil energy reserves in the U.S. are coal.)

8. It is the stated policy of Japan, the European common market, Australia, and the United States to reduce their dependence on oil through the increased use of coal or coal-based fuel.

9. Perhaps no single product holds the potential of future export growth for the United States as does clean coal over the next twenty-five years. (The U.S. and Australia are the only two nations with infrastructure and reserves adequate to export more than 100 MM/TPY of coal annually @ $100/T x 100MM = $10 billion annual export sales for each 100MM tons sold.)

10. A major increase in the export of conventional coal will require further massive investments in port, storage, and transportation facilities. (Formulation of clean coal as CWM obviates the need for much of this capital investment as the infrastructure which exists to support international liquid petroleum transport can be used.)

11. A number of major U.S. companies including Martin Marietta, Florida Power, Arco, and Babcox Wilcox, are financially committed to making coal slurry technology a reality.

12. Coal conversion technologies to synthetic fuels which require high temperature and pressure are not likely to be competitive with oil as long as the huge capital costs keep escalating for these giant projects more rapidly than conventional petroleum prices.

13. Physically cleaned coal (as opposed to chemically cleaned coal) has a practical limitation of approximately 1% ash for most coals and no capability for organic sulfur removal. In addition, it has some losses to tailings or a midling product. These constraints may preclude such material from most applications where petroleum products are now used.

14. In the area of chemical coal beneficiation, the only method known to substantially remove the ash from coal, without employing temperatures that drive off volatiles or otherwise alter the available hydrocarbons, is to leach with hydrofluoric (HF) and hydrochloric (HCl) acids.

15. The three criteria which must initially be met by any commercially viable alternative to petroleum are:

    A. Cost competiveness with petroleum products in a free market environment.

    B. Adequate fluidity to allow direct substitution (given minor modifications) in storage, transportation and combustion systems designed for petroleum use.

    C. Sufficiently pure hydrocarbons composition to meet or exceed environmental and equipment requirements.

16. Regardless of how coal is introduced to systems designed for petroleum use, ash must be reduced to near petroleum levels (probably below .5% as a minimum) to avoid fouling and other disfunctions of such systems.

Appendix C

POTENTIAL COMPETING PROCESSES

Molten alkalies will dissolve silicates which is the basis of TRW's Gravi-melt and G.E.'s Microwave processes being investigated by the D.O.E. Although these processes remove organic sulfur, they cause a loss of volatiles from the coal during contact with molton alkali and dissolve hydrocarbons during aqueous rinsing phase. Processing costs appear to be substantially higher than those for the Purged Carbons™ process.

Physical coal cleaning at best can only achieve about 1% ash for most coals. The Ostisca T process separates coal from an aqueous slurry of coal and mineral matter through use of trichloroflurohydrocarbon (freon). The process requires extreme fine grinding, to achieve near complete liberation (5 micron to 20 micron to size, depending upon the coal). This process is highly coal dependent. Only a few coals can be cleaned to less then 0.3% ash, and Western (Subbituminous) coals cannot be beneficiated by the process. From available information, the costs of this process also appear to be higher than for Purged Carbons™.

Appendix D

TABLE 1

PROJECT DEVELOPMENT STAGES

Stage	Development	Major Activities	Scale of Operation	Variability In The Slated Economics	Duration (Months)	Cost $000,000
I.	Bench	Process development, patent work, economics	75 lbs batches	± 30%	24 (completed)	2
II.	Continuous	Continuous operation, data acquisition, patenting efforts, product for testing economics	3 tons/day (250 lbs/hr)	± 20%	18	7
III.	Demonstration	Continuous operation engineering data and verification, product for testing economics	120 tons/day (40,000/TPY)	± 10%	18	50
IV.	Commercial	Engineering, construction start-up production	1 - to 5,000,000/TPY	--	30	350

Appendix E

TABLE 2

ONE YEAR BUDGET

for

PURGED CARBONS™ DEVELOPMENT, CONTINUOUS - STAGE II

Item	$000
Technical	
Pilot Plant, Capital Cost (6 months)	$ 1,200
Pilot Plant, Operating Cost (6 months)	650
Process Economics	200
Laboratory Program (12 months)	780
Consultants, Including Engineering Company	300
Patents, Legal	100
Transportation	300
Utilization	1,000
Subtotal	$ 4,530
Contingency (15%)	635
	$ 5,165
Operational	
G&A	835
	$ 6,000

NOXSO

THE NOXSO PROCESS for the Simultaneous Removal of SO_2 and NO_x from Combustion Flue Gas

The NOXSO Process is a dry, regenerable process for the removal of both SO_2 and NO_x from combustion flue gas. The process was developed over several years of laboratory research conducted by Princeton Chemical Research Company. NOXSO Corporation has purchased all rights to the process and is currently conducting an extensive program aimed at its commercialization.

This document provides a summary of information about the NOXSO process and the test program being conducted to determine its commercial potential. The following sections describes briefly the process and its key features, the prepilot program currently underway and gives pertinent results to date, and the proposed future development work.

In September of 1979, NOXSO Corporation was formed with an initial capitalization of $400. The Company's objectives were to purchase the NOXSO Process and to file a registration statement with the Securities and Exchange Commission to permit the sale of the Company's common stock. The aim of the initial public offering was to raise sufficient funds for the purchase of the NOXSO Process and to finance the small-scale testing of the process at a coal-fired power plant. The Company's public offering was completed in October, 1980 with the sale of stock to about 300 shareholders throughout the country. In November of 1980 the Company commenced operations and exercised its purchase option on the NOXSO Process for $200,000 plus future payments based on profits derived from the process.

During the period from November, 1980 through December, 1981, the Company pursued the design and assembly of the prepilot test system. Critical agreements were executed with

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2625 H.C. Mathis Drive
Paducah, KY 42001

(502) 444-6474

28-916 0 - 84 - 78

W.R. Grace for supply of sorbent and technical support, at no cost to NOXSO, the EPA for loan of an SO_2/NO_x monitoring system, and TVA for permission to test at Shawnee Steam Plant (see NOTE below). In January of 1982 the pre-pilot test system was shipped to Paducah, Kentucky, for installation and testing.

In September, 1982, the Department of Energy awarded NOXSO a contract for $313,000 to continue its testing program through July, 1983. It is expected that all the goals of the pre-pilot test program will be accomplished at that time.

NOTE: The NOXSO pre-pilot test is not sponsored by the Tennessee Valley Authority and the presence of the system at the Shawnee Steam Plant should not be construed as an endorsement of the process by TVA.

THE PROCESS

A schematic diagram illustrating the flow diagram of a commercial scale NOXSO air pollution control system installed on a coal-fired utility boiler is shown in Figure 1. The following discussion covers the key components of the NOXSO system: the sorption step, the regeneration step, and the NOXSO sorbent.

The NOXSO Process utilizes a solid, granular sorbent to capture both SO_2 and NO_x contained in coal-fired power plant flue gas.* The sorption is carried out at normal utility boiler post-air heater temperatures (240 to 300^{o}F) and, by virtue of the use of a moving bed, fluid bed, or dispersed phase reactor, there is no need for high efficiency particulate collection prior to the sorption vessel. Since the flue gas is not cooled during the cleaning steps, flue gas reheat is not necessary. The sorption step of the NOXSO Process was extensively tested at the laboratory scale in the early 1970's, with more than 140 sorption/regeneration cycles completed on the optimized sorbent formulation.

The NOXSO sorbent, once loaded with SO_2 and NO_x as sulfates and nitrates, can be restored to its active state by treatment with H_2S at elevated temperatures. Under proper conditions of temperature, residence time, and H_2S concentration, the products of regeneration are elemental sulfur and nitrogen. The regenerated sorbent is returned to the sorption vessel. A portion of the sulfur is treated with a reducing gas to provide a stream of H_2S for the regeneration step.

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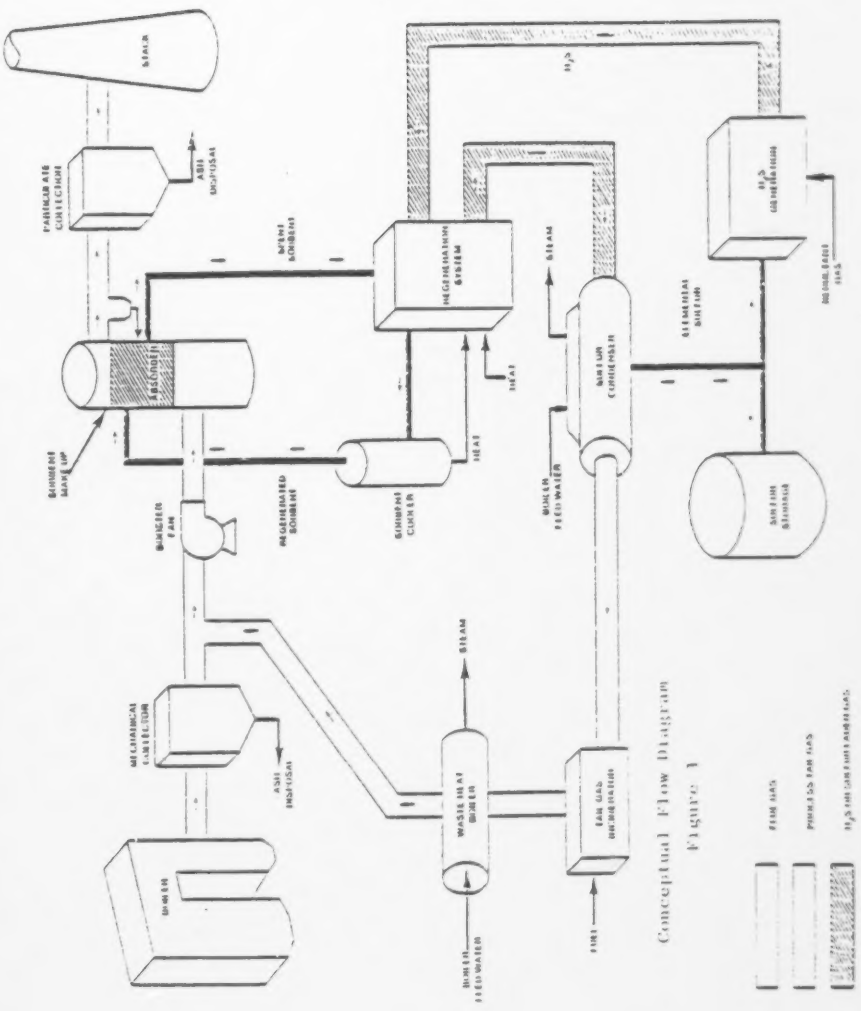
* SO_2 (sulfur dioxide) and NO_x (nitrogen oxides) are believed to be the major precursors of acid rain.

NOXSO



Conceptual Flow Diagram
Figure 1

The chemical reactions occurring in the adsorption and regeneration cycle are rather complicated. The analytical and test results to date indicate the following reactions are the most likely chemical reactions involving in the process:

Sorbent Preparation

$$Na_2CO_3\ (s) + Al_2O_3\ (s) \longrightarrow 2NaAlO_2\ (s) + CO_2\ (g)$$

Adsorption

$$2NaAlO_2\ (s) + SO_2\ (g) \longrightarrow Na_2SO_3\ (s) + Al_2O_3\ (s)$$

$$2NaAlO_2\ (s) + SO_2\ (g) + \tfrac{1}{2}O_2\ (g) \longrightarrow Na_2SO_4\ (s) + Al_2O_3\ (s)$$

$$2NaAlO_2\ (s) + NO_x\ (g) + (5/2 - x)O_2\ (g) \longrightarrow 2NaNO_3\ (s) + Al_2O_3\ (s)$$

Regeneration

$$Na_2SO_3 + Al_2O_3 + 2H_2S \longrightarrow 2NaAlO_2 + 3S + 2H_2O$$

$$Na_2SO_4 + Al_2O_3 + 3H_2S \longrightarrow 2NaAlO_2 + 4S + 3H_2O$$

$$2NaNO_3 + Al_2O_3 + 5H_2S \longrightarrow 2NaAlO_2 + N_2 + 5S + 5H_2O$$

The original developer of the NOXSO Process, Princeton Chemical Research, screened dozens of sorbent formulations before selecting sodium oxide on an alumina substrate for detailed optimization and life testing. It is important to note that the production of NOXSO sorbent does not present any technological problems in that it is produced from readily available materials (commercial alumina granules and sodium carbonate) by application of processes and equipment widely used in the catalyst industry. W.R. Graces's Davison Chemicals Division provided the sorbent presently being used in the ongoing pre-pilot test.

The NOXSO Process patent No. 4,323,544; entitled "Process and Sorbent System for Removing Nitrogen and Other Oxides from Gas Streams" was issued on April 6, 1982. The Company is continuing to pursue an additional application which contains claims regarding the NOXSO sorbent.

The NOXSO Process exhibits the following important features and advantages:

- The scrbent captures both SO_2 and NO_x, with removal efficiencies in excess of 90 percent being readily achievable.
- The process is dry and cleaned flue gas does not require reheat prior to release. The system will be ideally suited for retrofit applications since the flue gas is treated at normal utility boiler post-air preheater temperatures.
- When equipped with a baghouse for final particulate cleanup, the NOXSO Process will meet the NSPS for SO_2, NO_x, and particulate matter from coal-fired electric generating plants.
- The sorbent, sodium oxide on alumina, is inexpensive and does not contain potentially hazardous heavy metals.
- Recovered sulfur is in the form of marketable elemental sulfur. No sludge is generated.
- NO_x is converted to molecular nitrogen and released to the atmosphere.
- Ammonia is not required as a feedstock, as is the case with conventional Selective Catalytic Reduction NO_x removal processes.
- Water pollution is not a problem in the NOXSO Process, as is the case with the wet SO_2/NO_x systems in use in Japan.

NOXSO

THE PRE-PILOT TEST

Experience has shown that the progression from laboratory-scale testing with synthetic flue gas to small-scale field testing with actual flue gas constitutes a critical, and often fatal, step in the development life of pollution control technologies. Recognizing the importance of this step, NOXSO Corporation has devoted its first efforts and resources toward the accomplishment of this phase of testing. To date, more than twelve months of testing has been completed and the NOXSO Process has graduated from examination of technical feasibility to optimization of technical and economic factors and extended life testing of the NOXSO sorbent.

The NOXSO test facility is housed in a mobile trailer and requires only electricity and flue gas from the power plant. It is virtually self-contained. The facility contains two separate test rigs, both utilize flue gas in their operation. The smaller system treats 0.2 actual cubic feet per minute (ACFM) of flue gas and is used to study the process chemistry and kinetics. The pre-pilot system treats 60 ACFM of flue gas. NOXSO feels that this size operation represents an ideal prelude to a two megawatt Pilot Demonstration Unit.

The following are features and results of the ongoing pre-pilot test:

- To date more than 400 hours of on-stream absorption time has been accumulated. Pollutants removals have been 95-96% for SO_2 and 92-93% for NO_x.
- Approximately 80 successful sorbent regenerations have been carried out. The sorbent is consistantly regenerated to better than 95% of its initial capacity.

- Several sorbent formulations have been tested for chemically induced attrition. The material currently being used has shown no signs of being degraded by the sorbtion reaction at temperatures from 225 to 350°F.
- A series of endurance tests of the sorbent material is underway to determine the potential for long term chemical and physical degradation of the sorbent material.
- The process chemistry has been well established, kinetic data obtained for sorption and regeneration reactions, and the design equations derived.
- A key element of the test program is a conceptual design of a full scale (500 MWe) facility and a cost study being conducted by a NOXSO contractor. This work is approaching completion.

FUTURE DEVELOPMENT WORK

The present field tests were initially funded solely by private monies to October, 1982. A total of more than $900,000 were spent. The Department of Energy has provided support of the test program for the last eight months. It is expected that by the end of July (1983) the test program, including a preliminary economic study will be complete and the commercial promise of the NOXSO process established.

As the test efforts continue to show promise for the NOXSO Process, we anticipate a continuing development program leading to a first commercial application in the mid 1980's. The schedule for the continuing work will depend upon the availability of adequate financial support from government agencies, interested process users and/or investors. The next two steps in this program are as follows:

- o Pilot Demonstration Unit (PDU) Design and Test Plan Development--
  Starting in August, 1983 and continuing through January, 1984, NOXSO Corporation with W.R. Grace and an appropriate architect/engineer firm, would use output data from the present test to design a two megawatt pilot plant. Adjunct to this, laboratory work would continue as necessary to further define the process characteristics. Also, a site would be selected, construction cost estimated, and operation and test plans prepared.
  Estimated cost--$400,000

- o Pilot Demonstration Unit Construction and Operation--
  Starting in early 1984 and continuing for 21 months, a pilot unit would be built and operated. The schedule would include six months for construction, 3 months for shakedown and modification, and one year of operation.
  Budgetary Cost Estimate--$4-5 million

Further development steps will depend upon the success of the PDU. The need for an intermediate-size test prior to a first commercial installation will depend to some extent upon the urgency of the need for simultaneous control as determined by the scientific and regulatory communities.

WRITTEN STATEMENT

OF THE

AMERICAN GAS ASSOCIATION

BEFORE JOINT HEARINGS OF THE

SUBCOMMITTEE ON ENERGY DEVELOPMENT AND APPLICATIONS

AND THE

SUBCOMMITTEE ON NATURAL RESOURCES, AGRICULTURE RESEARCH AND ENVIRONMENT

OF THE

HOUSE COMMITTEE ON SCIENCE AND TECHNOLOGY

ON ACID RAIN: IMPLICATIONS FOR FOSSIL ENERGY R&D

September 20, 1983

- -

Mr. Chairman and members of the Committee:

The American Gas Association is a national trade association composed of nearly 300 natural gas distribution and transmission companies serving over 160 million U.S. consumers. On behalf of both our members and our consumers we would like to put before this Committee an easily implemented and economical technique of reducing sulfur oxide air pollution. Our strategy is called the Select Use of natural gas. It involves mixing natural gas -- which emits virtually no sulfur oxides when burned -- with more polluting fuels, such as coal or high sulfur oil. Clean burning natural gas offsets or reduces the amount of air pollution that would have been emitted if just coal or high sulfur oil were

burned. This reduction is important since sulfur oxide emissions are reported to be causes of acid precipitation.

Many bills have been introduced in both the House and the Senate to control acid rain. Some of these bills require specific control strategies, typically flue gas desulfurization, i.e., scrubbers. While we do not denigrate the goals of any of these bills, we disagree strongly with those that require a single solution. We believe that:

(1) Acid rain reductions should be achieved with performance standards, leaving the plant operator free to choose the most efficient control strategy; and

(2) Calculations that purport to show that acid rain controls will cost X or Y, and that are based on a single control strategy, are not useful and can even be misleading.

The most efficient control strategy for a particular plant depends on factors at that plant. As long as environmental goals are met, it makes sense to let the plant operator choose the control strategy that best fits the plant. Select use of gas, low sulfur coal blending, coal washing, scrubbers, and other strategies each have a place. We should consider and encourage various ways to reduce acid deposition.

What Is Select Use?: An Explanation of the Strategy

Natural gas is an extremely clean burning fuel. It emits virtually no sulfur oxides or particulates when burned. Its carbon dioxide, carbon monoxide, nitrogen oxide, and non-methane hydrocarbon emissions are all lower than oil or coal. For example, a large gas boiler (either industrial or power plant) on

average emits only .0006 pounds of SO_2 per MMBtus, while a boiler on an average grade of residual fuel oil (1.5% sulfur) emits 1.6 lbs SO_2/MMBtus. A similar boiler operating on 2% sulfur coal emits 3.4 lbs/SO_2/MMBtus. In a well-known case, the Narragensett Electric Company in Rhode Island used the clean burning attributes of natural gas to offset emissions from a switch to a higher sulfur oil. The utility operated two boilers on expensive low sulfur oil. In order to reduce fuel costs, it switched one boiler to high sulfur oil and the other to natural gas. By converting a single boiler to gas, the company was able to offset the increased SO_2 from the high sulfur oil and still comply with the area's strict SO_2 limits. This same strategy can be used with Eastern high sulfur or Western low sulfur coal. The following charts show the amount of natural gas (in percentages) that could be used with coal or oil, with varying sulfur contents. The percentages vary, of course, with the stringency of local limits on SO_2 emissions (shown on the left axis).

The data on the following pages show the significant environmental benefits of using natural gas.

A1

APPENDIX 1[1]

PERCENTAGE OF FUEL MIX REQUIRED TO BE GAS FOR VARIOUS GRADES OF COAL AND SO_2 EMISSION LEVELS

SO_2 SIP (lb./MMBtu)	Sulfur Content of Eastern Coal[2]				Sulfur Content of Western Coal[3]			
	.8%	1%	2%	3%	.8%	1%	2%	3%
0.6	53%	62%	81%	87%	61%	68%	84%	89%
0.8	37	49	75	83	47	58	79	86
1.0	22	37	68	79	34	47	74	82
1.2	6	24	62	75	21	37	68	79
1.4	–	11	56	71	8	26	63	75
1.6	–	–	50	66	–	16	58	72
1.8	–	–	43	62	–	5	53	68
2.0	–	–	37	58	–	–	47	65
2.2	–	–	31	54	–	–	42	61
2.4	–	–	24	49	–	–	37	58
2.6	–	–	18	45	–	–	32	54
2.8	–	–	12	41	–	–	26	51
3.0	–	–	5	37	–	–	21	47

[1] *Select Gas Use for Environmental Purposes*, American Gas Association 20 (January, 1983).

[2] 12,000 Btu/lb.

[3] 10,000 Btu/lb.

APPENDIX 2[1]

PERCENTAGE OF FUEL MIX REQUIRED TO BE GAS FOR[1] VARIOUS GRADES OF RESIDUAL OIL AND SO_2 EMISSION LEVELS

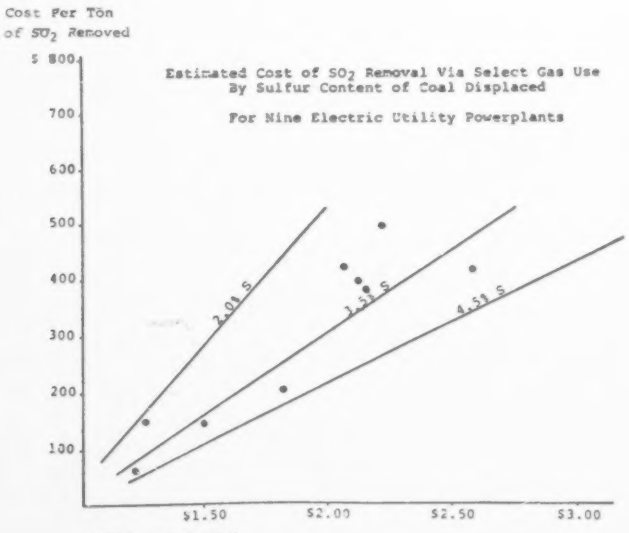
SO_2 SIP (lb./MMBtu	Sulfur Content of Oil				
	.25%	.5%	1%	2%	3%
0.2	23%	62%	81%	90%	94%
0.4	–	23	62	81	87
0.6	–	–	43	71	81
0.8	–	–	24	62	75
1.0	–	–	5	52	68
1.2	–	–	–	43	62
1.4	–	–	–	33	56
1.6	–	–	–	24	49
1.8	–	–	–	14	43
2.0	–	–	–	5	37
2.2	–	–	–	–	30
2.4	–	–	–	–	24
2.6	–	–	–	–	17
2.8	–	–	–	–	11
3.0	–	–	–	–	5

[1] *Select Gas Use For Environmental Purposes*, American Gas Association 21 (January, 1983).

Choosing the Most Efficient Acid Rain Control Strategy: The Benefits of Natural Gas

With select use of gas, the cost of sulfur oxide reductions can vary from $50/ton of SO_2 removed to over $1000/ton. This range of costs is not unusual. A study by ICF, Inc. a consulting firm, postulated a range of control costs for fuel switching (high to low sulfur coal or oil) and scrubbing from $250 - $2000/ton. When one considers that many select use options fall in the $200 - $400/ton range, the reader can immediately see that gas and coal or high sulfur oil mixtures are economical alternatives to scrubbing or fuel switching.

The following chart graphically displays the site specific benefits of select use at nine powerplants.



1/ Cost of Gas - Cost of Coal

Note: Assumes O&M of coal-fired utility boiler exceeds gas boiler O&M by $1.09/mmBtu based on information contained in: Thermal-Electric Plant Construction Cost and Annual Production Expenses, DOE/EIA.

●- Represents actual plant operating data (cost of gas, cost of coal, sulfur content of coal) for December, 1982. See: Cost and Quality of Fuels for Electric Utility Plants, DOE/EIA.

For completeness, the ICF $250 - $2000 cost range for non-select use control options is:

Representative Annualized Costs of SO2 Reduction Alternatives*

Control Strategy	$/ton SO2 Removal (mid-1982 dollars)
Shift from High- to Low-Sulfur Coal**	$ 250 - $ 300
Shift from High- to Medium-Sulfur Coal**	350 - 400
Shift from Medium- to Low-Sulfur Coal**	400 - 450
Shift from High- to Low-Sulffur Residual Oil	450 - 600
Shift from Unscrubbed to Scrubbed High Sulfur Coal***	400 - 500
Shift from Unscrubbed to Scrubbed Midium-Sulfur Coal***	600 - 1,100
Shift from Unscrubbed to Scrubbed Low-Sulfur Coal***	1,200 - 2,000

*Cost will vary considerably, depending upon region and plant size. Costs per ton presented in this table are intended to be representative of average control costs only.

**Costs include upgrading particulate removal equipment

***Based on retrofit scrubbing costs for a 500-megawatt powerplant.

Source: ICF, Incorporated

We think, however, that ICF may have understated the costs of such a massive switch to low sulfur coal. For example, the model tends to understate the costs of increased rail transportation. Also, the other effects of fuel switching need to be considered. These include unemployment in Eastern coal mines, plant derating, and increased solid waste disposal.

A Comparison of Costs: Select Use and Other Options

In many situations, but not in all or even necessarily most, select use can be the most economical control option. Given that the best control option for a specific site depends on the

particular characteristics of that site, we can roughly compare select use with other options using the following hypothetical case:

SELECT USE	If 50 large (750 MW each) coal-fired electric plants were burning 2.5 percent sulfur coal and emitting 5 million tons of SO_2 every year, and if our goal were to reduce these emissions by 30% (1.5 million tons/year), the theoretical cost, employing select use of gas with coal, would be $300 - $600 million, in 1982 dollars.
LOW SULFUR COAL/ SCRUBBERS	Using the same assumptions, the cost of using low sulfur coal and scrubbers would be $1.8 - 3 billion.
HIGH SULFUR COAL/ SCRUBBERS	Again using the same assumptions, the cost of using high sulfur coal and scrubbers would be $600 - 750 million.
LOW SULFUR COAL ONLY	Lastly, with the same assumptions, the cost of a complete conversion to low sulfur coal would be $375 - 450 million.

Thus, Select Use falls at the low end of the scale for acid rain control strategies. In addition to the quantifiable benefits of Select Use, we should also mention its intangible benefits. For example, Select Use is a flexible control strategy that can be combined easily with other technologies. In our 50 plant hypothetical, Select Use and low cost coal washing could reduce SO_2 by 50% or more -- well above our hypothetical goal of 30% -- while continuing to use 70%+ Eastern high sulfur coal. This is in sharp contrast to low sulfur coal fuel switching which could cut Eastern coal consumption to 25%, in the worst case. In fact, we have prepared a short attachment which shows that Select Use can help Eastern high sulfur coal to retain a market.

Although the immediately preceding pages have been devoted to Select Use with coal, natural gas mixtures are also an excellent way to lower high sulfur oil emissions. There are hundreds of

power plants and industrial facilities that could use gas/oil mixtures either in adjacent boilers, or in the same boiler. Professor Alex Green of the Interdisciplinary Center for Aeronomy and Atmospheric Sciences at the University of Florida estimated in 1980 that consumers could save $12.5 billion per year in electricity costs by switching oil-fired powerplants to gas-coal mixtures. (While the Green work is becoming dated, the bottom line is still clear: the potential savings are large.)

Select Use Is a Feasible and Well-Proven Technology

A special advantage of Select Use is that it is a readily usable technique, with relatively low capital costs. Although much acid rain research is still ongoing, as a nation we may feel that we need near term controls. Thus, one possibility is that plant operators could employ Select Use on an interim basis, during the study period. This strategy avoids the long leadtimes and large investments of major control equipment, as well as the economic dislocations of high to low sulfur coal switching. It also preserves each of those options so that -- if research pinpoints acid rain sources -- we can use our resources efficiently for the best controls for that source.

A.G.A. recently compiled a list of facilities that use some mixture of natural gas and another fuel. This tabulation shows that Select Use is a control strategy that we can use here and now.

9/2/83

Cases of Natural Gas Used in Combination
With Other Fuels

Type of Gas Use	State	User	Gas Company Supplier
		TRUE SELECT USE APPLICATIONS	
E,X	RI	Narragansett Electric Co.	The Providence Gas Company
E,X	VT	Burlington Electric Co.	Vermont Gas Systems, Inc.
E,X	CT	Uniroyal	Connecticut Light & Power Co. (Subsidiary of Northeast Utilities Co.)
E,X	NJ	Union Carbide	Public Service Electric & Gas Co.
E,X	NJ	Public Service Electric & Gas Co.	Public Service Electric & Gas Co.
E,X	PA	Bellefield Boilers	The Peoples Natural Gas Co.
		OTHER APPLICATIONS OF GAS IN COMBINATION WITH OTHER FUELS	
I	PA	Scott Paper	Suburban Gas Co.
F,O	NJ	Atlantic City Electric Co.	South Jersey Gas Co.
O	MO	James River Power Plant	Northwest Central Pipeline Co. (formerly Cities Service Gas Co.)
F	WVA	FMC Corporation	Cabot Corporation Ajax Pipeline Co.
		COMBINATION FUEL CAPABLE FACILITIES	
MFC(F,O)	AZ	Tucson Electric Co. (Presently under conversion; company currently plans to limit gas use to flame stabilization and operational uses but would have technology for full "select use")	Southwest Gas Corp.
DFC	MA	Boston Edison Co.	Boston Gas Co. (Subsidiary of Eastern Gas & Fuel Associates)

LEGEND

X - Select-Use of Gas (i.e., the combustion of gas, in conjunction with less environmentally attractive fuels, has the effect of significantly reducing emissions or preventing emission increases that would otherwise occur)

E - Although the use of gas has significant environmental benefits, the company does not consider itself irrevocably obligated to continue "select use" and reserves the right to alter or abandon its "select use" practices if the economics of "select use" become unfavorable.

I - Gas is Used for Ignition and Start-up Purposes Only

O - Gas is Used to Reduce Operating Problems

F - Gas is Used for Flame Stabilization

MFC - Can burn gas, oil or coal - or combinations thereof

DFC - Can burn gas or oil - or combinations thereof

If plant operators can choose Select Use, as one of many options, as much as 1 Tcf/year of natural gas might ultimately be used for environmental compliance. Nationally, the natural gas industry is quite capable of supplying this market. 1 Tcf/year is only approximately 5% of current or projected yearly U.S. production. At the present time, we estimate that the U.S. has as much as 3-4 Tcf excess deliverability: that is, gas shut-in or not taken. A 1 Tcf investment could translate into a 2 million ton/yr. SO_2 reduction. By combining Select Use with coal washing, that same 1 Tcf could reduce SO2 by nearly 4 million tons/year. If we limit ourselves to our 50 power plant hypothetical, 650 Bcf/year (just 3.6% of current U.S. production and less than a quarter of the surplus deliverability now at hand) could reduce emissions by 1.5 M tons/yr., or 2.5 M tons/yr. with coal washing.

Summary

The benefits of multiple control strategies for acid rain are many. Select Use, in particular:

- Maximizes the use of Eastern coal and minimizes the economic and social dislocations among mining communities;
- Avoids plant derating, increased solid waste, increased particulates, and railroad transportation problems associated with straight coal strategies;
- Offers prompt emission reductions since the equipment is proved and readily available;
- Can buy the nation valuable time since Select Use can be used temporarily to reduce emissions quickly while research is ongoing; and
- Avoids the high capital costs of a mandatory scrubber program.

With myriad benefits, Select Use is a vital alternative for the nation to consider. Now is the time to take advantage of the environmental benefits of our cleanest burning fossil fuel. We can do that by: (1) repealing incremental pricing and the Fuel Use Act gas and oil use restrictions; (2) ensuring that future acid rain legislation does mandate performance standards rather than specific fuels or control strategies; and (3) urging the Environmental Protection Agency to accept Select Use as a control technology under existing provisions of the Clean Air Act.

ALCALDE, HENDERSON & O'BANNON

GOVERNMENT & PUBLIC AFFAIRS CONSULTANTS

September 29, 1983

RECEIVED

OCT - 4 1983

COMMITTEE ON SCIENCE
AND TECHNOLOGY

Honorable Don Fuqua, Chairman
Science and Technology Committee
U.S. House of Representatives
2269 Rayburn House Office Building
Washington, DC 20515

Dear Don:

I am writing to request that the attached study from ICF, Inc. prepared for the Alliance for Clean Energy (ACE), be submitted as part of the record for the September 20 Joint Hearing on Acid Rain before the Subcommittee on Energy Development and the Subcommittee on Natural Resources.

In examining the societal effects of various acid rain control strategies, I believe the attached ICF study sheds some light on the impact acid rain controls could have on miner employment.

In this study, ICF compared H.R. 3400 (the Waxman/Sikorski Acid Rain bill) with a cost-effective program that would produce equivalent reductions in sulfur dioxide emissions. ICF concluded that the cost effective equivalent will have little effect on mining employment. As a matter of fact, the study found:

- Coal mining employment goes up 54% over 1980 levels -- this creates 120,200 new mining jobs.
- 21 coal producing states both East and West gain 138,100 jobs by 1995
- Employment declines in 4 states -- but 98% of the declines are in just 2 states (Ohio and Illinois).
- Cost-effective acid rain controls actually add new jobs to several states -- e.g., West Virginia gains 11,000 more

The study also found that H.R. 3400, would cost consumers and taxpayers enormous sums. Protecting high-sulfur coal producers by requiring mandatory scrubbers would cause these results:

- Capital costs increase fourteenfold to $21.5 billion by 1995.

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- Annual costs will be 46% greater.
- Total mining employment will probably be lower under H.R. 3400 - than under the cost-effective equivalent.
- Requiring scrubbers produces no net gains in environmental benefits despite the huge increase in costs.

As Mr. Jan Mares, Assistant Secretary for Fossil Energy, Department of Energy pointed out in testimony presented to the Subcommittees during the hearings, new coal technologies are emerging. In the next ten years, many of these technologies will be commercially available. If we spend between $19 and and $21 billion in capital costs for scrubbers, there will be very little funding left for newer, more cost effective technologies. In addition, scrubbers may be outdated in ten years.

ACE believes that electric utilities should have the freedom to choose the most cost effective compliance strategy for controlling acid deposition. It seems clear that attempts by Congress to mandate specific technologies would be an ineffective way to control the acid rain problem.

Thank you for allowing the Alliance for Clean Energy this opportunity to submit testimony as part of the record.

With best personal regards, I am

Sincerely,

Hector Alcalde

HA/c
Encl. ICF, Inc.

ICF INCORPORATED International Square, 1850 K Street, Northwest, Washington, D.C. 20006 (202) 862-1100

September 22, 1983

MEMORANDUM

TO : Alliance for Clean Energy

FROM : Daniel E. Klein

SUBJECT: Analysis of Coal Production and Employment Impacts of the Waxman-Sikorski Bill and a Cost-Effective Equivalent

This memorandum presents ICF's findings of an analysis of the Waxman-Sikorski Bill and a cost-effective equivalent. Special attention has been devoted to shifts in regional coal production and changes in employment in coal mining and transportation.

EXECUTIVE SUMMARY OF FINDINGS

- Three scenarios were examined for a 1995 forecast period. The base case represents a reference case forecast that assumes no acid rain mitigation legislation. The Waxman-Sikorski case is our analysis of H.R. 3400, and represents a reduction of about 9.5 million tons of sulfur dioxide emissions by electric utilities and industrial coal-burning facilities. The cost-effective equivalent case was designed to result in the same level of sulfur dioxide emission reductions as achieved in the Waxman-Sikorski case, but permitted states and utilities free choice to pursue emission reduction strategies in the most cost-effective manner.

- Costs -- The Waxman-Sikorski case results in higher costs to the nation than the cost-equivalent case. The requirements in Waxman-Sikorski would increase the net cost to the nation by about one billion dollars per year (in mid-1982 dollars) relative to the cost-effective equivalent. However, the direct costs to utilities would be significantly less due to subsidy payments.

- Regional coal production -- In the cost-effective equivalent case, fuel switching is often selected as a least-cost means of reducing sulfur dioxide emissions. Accordingly, forecasted coal production tends to decline in higher-sulfur regions such as Northern Appalachia and the Midwest, and increase in lower-sulfur regions such Central Appalachia and the Rockies. Under the Waxman-Sikorski case, there would be a heavy reliance upon retrofit scrubbers as the means for reducing emissions. As such, there are relatively few shifts in coal production relative to base case forecasts, with some regions actually showing an increase in forecasted high-sulfur coal production.

- Employment shifts -- Relative to the Waxman-Sikorski case, the cost-effective equivalent case could result in many jobs shifting from higher-sulfur to lower-sulfur regions by 1995. Relative to 1980 employment levels, the cost-effective equivalent case indicates lower employment levels in higher-sulfur regions such as the Midwest and parts of Northern Appalachia. For the United States as a whole, the forecasted number of jobs in coal mining and transportation shows only modest changes in the three 1995 cases, but appears to be slightly higher in the cost-effective equivalent case than under Waxman-Sikorski.

FRAMEWORK FOR ANALYSES

Cases Examined

In order to evaluate the potential magnitude of the changes in the coal market, we have analyzed three scenarios for 1995:

- The base case represents a reference case forecast that assumes no acid rain mitigation legislation. This is the same base case that was specified by EPA and used in recent acid rain analyses.

- The Waxman-Sikorski case is an analysis of HR-3400 which was performed for EPA, and includes its provisions for mandatory scrubbing at selected plants, scrubber subsidies, sulfur dioxide emission rollbacks, and other requirements specific to this particular bill. In this case, the total emissions of sulfur dioxide by electric utilities in the U.S. is forecasted to be 10.7 million tons in 1995, a reduction of about 9.0 million tons from the 1995 base case. Industrial coal-burning facilities were assumed to reduce sulfur dioxide emissions by about 0.5 million tons through fuel shifting.

- The cost-effective equivalent case was designed to result in the same level of sulfur dioxide emissions in the U.S. as achieved in the Waxman-Sikorski case. However, this case did not provide scrubber subsidies, and did not require specific sulfur dioxide control strategies or emission limits. States and utilities were free to pursue strategies for achieving the required sulfur dioxide reductions in the most cost-effective manner including retrofitting scrubbers, dispatching plants in a least-emissions manner, switching to lower-sulfur fuels, coal blending, and trading of emissions among states. As in the Waxman-Sikorski case, industrial facilities were assumed to reduce emissions through shifts to lower-sulfur fuels.

The Base Case and alternative cases are consistent with previous ICF analyses for EPA. The regional coal production levels are sensitive not only to the acid rain legislation assumptions, but also to the base case assumptions. The base case, initially specified by EPA, may overstate high-sulfur coal production. If so, then production and job losses (from 1980 levels) in high-sulfur regions resulting from acid rain legislation could be understated.

Coal Mining Employment

This analysis also incorporates an improved methodology for estimating employment changes in coal mining. Previous analyses of coal mining employment have relied upon use of historic productivity estimates applied to forecasted regional coal production.

However, the average productivity in 1980 includes production from many metallurgical coal mines which typically have lower productivity than those mines producing steam coals for the industrial and utility markets. Growth in coal demand, as well as shifts resulting from acid rain legislation, will be concentrated in steam coal production. Accordingly, in regions where metallurgical coal production is substantial, use of historic productivity estimates will tend to understate future productivity, overstate future employment levels, and exaggerate shifts (both gains and losses) in mining employment across the scenarios. Further, future productivity in a region could change from historic productivity as a result of changes in the proportion of surface and deep-mined coal, changes in mining technologies, changes in labor practices, and changes resulting from depletion of the more efficient mines.

In recognition of these problems inherent with the use of historic productivity values, we have undertaken the development of an improved methodology for estimating coal mining employment. Specifically, this methodology incorporates the same estimates of coal mining productivity that are used in ICF's Coal and Electric Utilities Model (CEUM) to forecast regional coal production and utility fuel use. In CEUM, coal supply is

represented in the form of coal supply curves. There are over 200 coal supply curves, one for each type of coal in each of 40 coal supply regions. Each supply curve is represented as a step function, where each step represents a different type of mine (in terms of mine size, seam thickness, overburden ratio, and other factors). In developing the minimum acceptable selling price for each mine-type on each coal supply curve, the mine costing program in CEUM develops an estimate of productivity in terms of tons per man-day. Using these productivity estimates, we estimated the number of workers associated with the forecasted coal production of any given coal-type and region. In so doing, this improved methodology is able to circumvent many of the problems inherent in the use of historic productivity values.

Coal Transportation Employment

This analysis has also incorporated improvements in the estimation of coal transportstion employment. In earlier analyses, changes in coal transportation employment were approximated by the changes in total ton-miles divided by a transportation productivity equal to that of a hypothetical unit coal train.

In this analysis, we have developed estimates of the labor productivity in coal transportation for each mode of transportation represented in CEUM. For example, transportation productivity for water movements of coal were estimated for 13 inland waterways, the Gulf intracoastal waterway, Great Lakes movements, and trans-oceanic movements. These productivity estimates were developed as functions of representative ship or barge size, crew size, average days per worker-year, and average speed. Similar methodologies were used in the estimation of labor productivity in unit train, single-car, and truck movements. In all cases, we have tended to be conservative (i.e., tending toward the low end of the range) in our estimates of the number of transportation workers involved; in particular, we have focused primarily upon those workers actually in motion with the coal and have tended to disregard support labor.

The resulting estimates of transportation labor productivity developed for this analysis can be summarized as follows:

	Ton-Miles Per Worker-Year (millions)	Workers per 10^9 Ton-Miles
Unit Train	53	19
Single-Car Rail	26	38
Trucks	0.6	1,695
Barges	7-52	19-153
Intracoastal Waterway	20-26	38-50
Great Lakes	17-44	23-58
Trans-Oceanic	97-243	4-10

These estimated worker requirements were used with the individual coal movements forecasted in CEUM. In CEUM, there is a transportation route description for coal moving between each origin and destination, by consuming sector. These route descriptions are comprised of mileages by mode of transportation. Using the productivity estimates summarized above, together with the route descriptions and forecasted coal flows, the number of coal transportation workers can be approximated for each forecast case.

POTENTIAL MARKET IMPACTS

The Waxman-Sikorski and cost-effective equivalent alternatives for implementing an acid rain mitigation strategy lead to substantial differences in forecasted retrofit scrubber capacity, coal consumption, regional coal production, total costs, and employment in coal mining and transportation. These changes are discussed below.

Scrubber Retrofits

Our analysis indicates that shifting to lower-sulfur fuels is often less expensive than retrofitting scrubbers on existing facilities as a means of reducing sulfur dioxide emissions. As such, there were only 2.5 gigawatts of retrofit scrubber capacity forecasted under the cost-effective equivalent relative to a 1995 base case without acid rain mitigation requirements. In contrast, the scrubber requirements and capital subsidy provisions of the Waxman-Sikorski case are forecasted to lead to 67.7 gigawatts of additional retrofit scrubber capacity in 1995. See Table 1.

TABLE 1

SCRUBBER CAPACITY UNDER ALTERNATIVE
ACID RAIN MITIGATION APPROACHES
(gigawatts)

	Scrubbed Capacity		Additional Retrofit Scrubbers in 1995	
	1980 Actual	1995 Base Case	Waxman-Sikorski	Cost-Effective Equivalent
Total U.S.	35.5	100.6	+67.7	+2.5

Changes in Coal Consumption

Coal's relative share of future energy requirements in the utility and industrial sector was assumed to be independent of any imposed acid rain regulatory program. Coal as a boiler fuel continues to remain more economic than oil and gas for many fuel applications within the utility and industrial sectors. Particularly in the electric utility sector, switching from coal to oil or gas is one of the least cost-effective emissions control strategies. Other factors such as regulatory, financial, managerial preference, and fuel distribution issues are likely to be more important in determining the

relative share of future energy markets for coal relative to oil and gas than will acid rain legislation. Therefore, the total amount of coal consumed by utility and industrial boilers was assumed to remain unchanged under the alternative acid rain mitigation scenarios.

However, the types of coal and oil demanded will shift significantly as some utilities and industrial facilities shift to lower-sulfur fuels to meet emission reduction requirements. Further, the demand for natural gas would likely increase under acid rain legislation because gas can be cost-effectively substituted for oil (though generally not for coal). However, this was not evaluated in this or any of the EPA analyses. Table 2 indicates the changes in forecasted 1995 coal consumption by sulfur level in the U.S. for the utility and industrial sectors.

TABLE 2

CHANGES IN FORECASTED 1995 COAL CONSUMPTION
IN THE U.S.
(10^{15} Btu (quads))

	1995 Base Case	Changes With Acid Rain Legislation: Waxman-Sikorski	Changes With Acid Rain Legislation: Cost-Effective Equivalent
Utility			
Low Sulfur	4.8	+0.2	+3.4
Low-Medium Sulfur	2.7	+0.5	+0.8
High-Medium Sulfur	6.2	-1.5	-1.6
High Sulfur	4.8	+0.9	-2.6
Total	18.5	+0.1	-
Industrial			
Low Sulfur	1.7	+0.4	+0.4
Low-Medium Sulfur	0.2	-0.1	-0.1
High-Medium Sulfur	0.8	-0.3	-0.3
High Sulfur	0.6	-0.1	-0.1
Total	3.3	-	-

Under the cost-effective equivalent case, the shift to lower-sulfur coals is more pronounced, reflecting the greater reliance upon fuel switching as a least-cost strategy. Under the Waxman-Sikorski case, the shift to lower-sulfur fuels is much less pronounced since the scrubber requirements and capital subsidies preclude or reduce the attractiveness of fuel switching. In fact, because of the large amount of retrofit scrubber capacity forecasted under the Waxman-Sikorski case, there would be a significant shift from high-medium sulfur coals to high-sulfur coals by some of the utilities retrofitting scrubbers.

Changes in Regional Coal Production

Consistent with the shifts to lower-sulfur coals by utility and industrial boilers, the cost-effective equivalent case would lead to a shift in the forecasted coal production by region. Table 3 shows coal production by aggregate region for 1995, and also indicates the changes from 1980 to 1995. Supply regions are shown in Figure 1.

Under the cost-effective equivalent case, higher-sulfur regions such as Northern Appalachia and the Midwest are forecasted to have lower levels of production than would have been the case in the absence of acid rain mitigation legislation. Our forecasts indicate 1995 production from these regions as being less than 1980 production levels. Conversely, lower-sulfur regions such as Central Appalachia and parts of the West are forecast to have significantly higher levels of coal production.

Under the Waxman-Sikorski case, there are only slight changes in forecasted coal production relative to the base case. This is because most of the sulfur dioxide emission reductions are attained through scrubber retrofits. As such, there is little shifting to lower-sulfur fuels in order to reduce emissions, and production in lower-sulfur producing regions such as Central Appalachia and parts of the West show little change from 1995 base case levels.

With the requirements and/or subsidies to retrofit scrubbers under the Waxman-Sikorski case, there is less incentive to pay a premium for lower-sulfur fuels at these plants. As a result, our forecasts under the Waxman-Sikorski case also indicate a shift within several of the higher-sulfur producing regions from high-medium sulfur coals to high and very-high sulfur coals.

For this analysis, few shifts to lower-rank coals were forecast. This reflects an assumption that individual boilers designed to burn bituminous coals would not find shifting to lower-rank coals economic after consideration of powerplant modification costs, capacity derates, heat rate increases, and increased costs for flame stabilization. To the extent that such shifts would occur, the estimated cost impacts would be lower, and the forecasted production increases in Eastern and Western bituminous low-sulfur coal fields would be less.

Total Costs

Relative to no legislation, both acid rain mitigation cases would result in higher costs to electric utilities and industry. See Table 4.

In the cost-effective equivalent case, cumulative capital costs by 1995 would increase by 1.5 billion dollars (in mid-1982 dollars) as a result of the acid rain legislation evaluated. This relatively small amount reflects the greater reliance upon fuel switching instead of retrofit scrubbing as a cost-effective means of reducing emissions. Under the Waxman-Sikorski case, the level of retrofit scrubber capacity is much greater. As a result, the cumulative capital costs exceed 21 billion dollars, most of which would be subsidized.

TABLE 3

REGIONAL COAL PRODUCTION[a]
(million tons)

Region	1980	1995			Change from 1980 to 1995		
		Base Case	Waxman-Sikorski	Cost-Effective Equivalent	Base Case	Waxman-Sikorski	Cost-Effective Equivalent
Northern Appalachia	185	213	202	157	28	17	(28)
Central Appalachia	233	295	299	350	62	67	117
Southern Appalachia	26	26	26	27	(1)	(0)	0
Total, Appalachia[b]	444	534	528	533	89	84	89
Midwest	134	182	187	88	48	53	(46)
Eastern Northern Great Plains	17	33	33	33	16	16	16
Western Northern Great Plains	125	250	250	270	125	126	145
Total, Northern Great Plains[b]	142	283	283	303	141	141	161
Central West	13	23	23	21	11	10	9
Gulf	29	126	126	126	97	97	97
Rockies	32	73	78	133	40	46	101
Southwest	29	61	59	78	32	30	49
Northwest	5	6	6	4	1	1	(1)
Alaska	1	12	12	12	12	12	12
Total, Rest of West[b]	109	302	305	375	193	196	266
Total U.S.[b]	830	1,300	1,303	1,299	470	473	469

[a] Regional details in Attachment A-1.

[b] Totals may not add due to independent rounding.

FIGURE 1

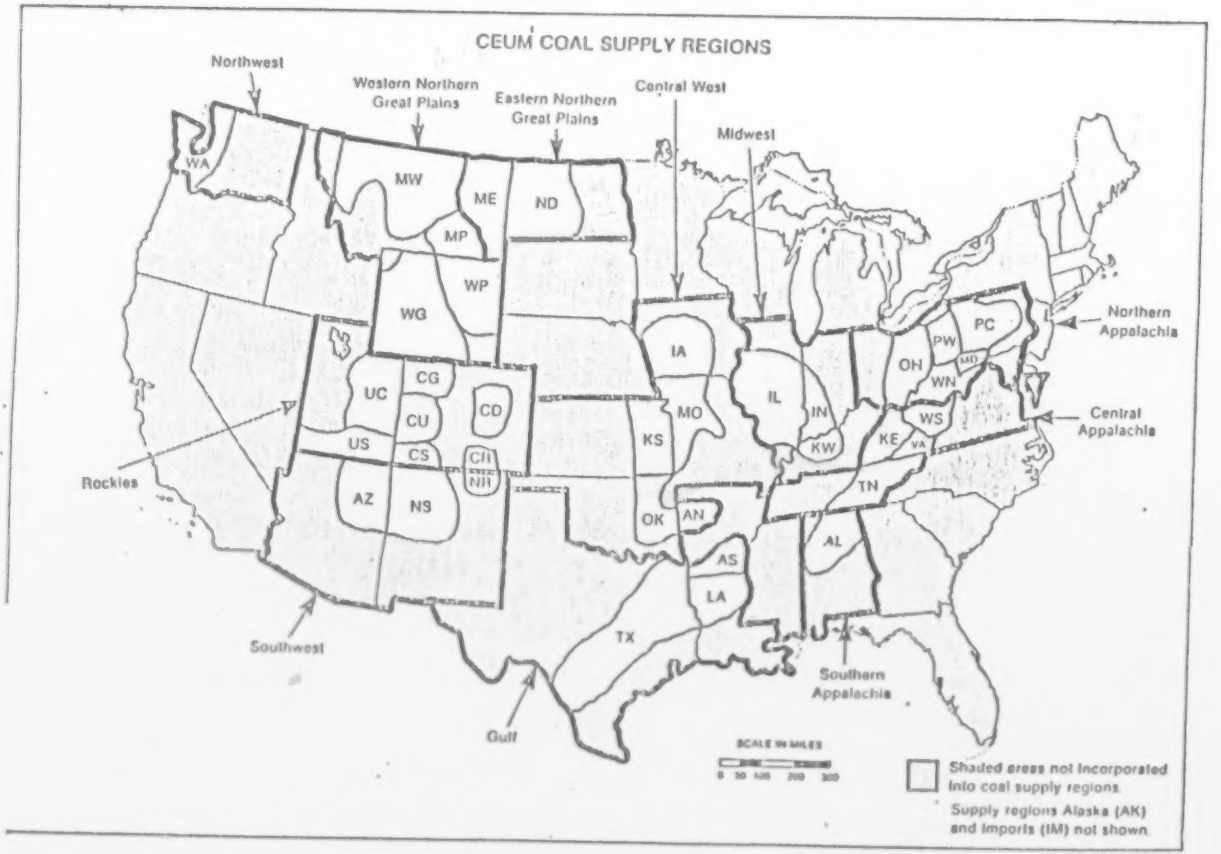
CEUM COAL SUPPLY REGIONS
Northwest
Western Northern Great Plains
Eastern Northern Great Plains
Central West
Midwest
Northern Appalachia
Central Appalachia
Rockies
Southwest
Gulf
Southern Appalachia
WA
MW
ME
ND
MP
WP
WG
UC
CG
CU
CD
US
CS
CN
NN
AZ
NS
IA
MO
KS
OK
AN
TX
LA
AS
IL
IN
KW
OH
PW
PC
MD
WN
WS
KE
VA
TN
AL
SCALE IN MILES
0 50 100 200 300
Shaded areas not incorporated into coal supply regions.
Supply regions Alaska (AK) and imports (IM) not shown.

TABLE 4

INCREASED CAPITAL AND ANNUALIZED COSTS IN 1995
DUE TO ACID RAIN LEGISLATION
(billions of 1982 $)

	Increases Relative to No Acid Rain Legislation	
	Waxman-Sikorski	Cost-Effective Equivalent
A. UTILITY SECTOR		
Cumulative Capital Costs		
Total	+21.5	+1.5
- Subsidy Payments*	+18.0	-
= Direct Cost to Utilities	+3.5	+1.5
Annualized Costs		
Total	+3.8	+2.6
- Subsidy Payments*	+2.1	-
= Direct Cost to Utilities	+1.7	+2.6
B. INDUSTRIAL SECTOR		
Annualized Costs		
Total	+0.4	+0.6
- Subsidy Payments*	-	-
= Direct Cost to Consumer	+0.4	+0.6
C. TOTAL		
Annualized Costs		
Total	+4.2	+3.2
- Subsidy Payments*	+2.1	-
= Direct Costs	+2.1	+3.2

* Location of where the subsidy costs are incurred cannot be determined independent of the subsidy financing scheme.

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On an annualized basis, the cost-effective equivalent case would entail total annual costs of 3.2 billion dollars (in mid-1982 dollars) relative to no acid rain legislation. The Waxman-Sikorski case would entail annual costs of 4.2 billion dollars (or one billion dollars more per year), half of which would be reflected in the subsidy.

There are a number of uncertainties with respect to the implementation of a subsidy program which could result in either higher or lower differential cost impacts. These uncertainties and the resulting impacts have not been addressed in this analysis.

Changes in Coal Mining and Transportation Employment

The potential shifts in regional coal production as a result of acid rain mitigation legislation could also lead to coal mining job losses in some regions, and gains in other regions. Additionally, coal distribution patterns would also change, leading to employment changes in the transportation industries.

Table 5 summarizes the changes in coal mining and transportation employment in 1995 under the different scenarios. Attachments A-1 and A-2 develop these estimates in greater detail.

-- Coal Mining

Because coal demand is forecasted to grow in all cases, 1995 coal mining employment is also forecasted to grow. Our estimates indicate that the net changes in coal mining employment in both the cost-effective equivalent and Waxman-Sikorski cases indicate a small decline from the forecasted base case national employment levels. The net national job losses under the Waxman-Sikorski case are slightly greater than those under the cost-effective equivalent case.

Consistent with the shifts in regional coal production, the cost-effective equivalent case could lead to shifts in regional coal mining employment. Under the cost-effective equivalent case, production and employment would decline from 1980 levels in higher-sulfur regions such as the Midwest and parts of Northern Appalachia. A forecasted decline is not necessarily the same as forced mined closings and/or layoffs, since between 1980 and 1995 some mines will have depleted their reserves and some miners will have retired.

Forecasted employment levels do not rise or fall proportional to changes in regional coal production, as had been the case in earlier preliminary analyses which relied upon the use of historic productivity estimates. For example, production in some regions (such as Colorado Uinta) is forecasted to rise at a much faster rate than employment levels. This is because much of the current production in these regions is comprised of lower productivity metallurgical mines, whereas the growth in production will largely be in higher productivity steam coal mines. In another example, intraregional shifts in coal production by sulfur content can lead to greater employment

TABLE 5

SHIFTS IN COAL MINING AND TRANSPORTATION EMPLOYMENT
IN 1995 UNDER DIFFERENT
ACID RAIN IMPLEMENTATION PROGRAMS

	Average Number of Workers in 1980 (10^3 Workers)	Change in Employment from 1980 to 1995 (10^3 Workers)		
		1995 Base Case	Waxman-Sikorski	Cost-Effectiv Equivalent
A. COAL MINING[a/]				
Northern Appalachia	69.1	+24.5	+17.0	+1.1
Central Appalachia	92.6	+41.3	+44.3	+68.2
Southern Appalachia	11.4	+2.9	+3.0	+3.7
Total, Appalachia	173.1	+68.7	+64.3	+73.1
Midwest	35.1	+17.0	+16.1	-10.1
Eastern Northern Great Plains	1.7	+1.3	+1.2	+1.3
Western Northern Great Plains	6.0	+6.8	+6.9	+8.8
Total, Northern Great Plains	7.7	+8.1	+8.1	+10.0
Central West	2.9	+5.2	+4.8	+4.6
Gulf	1.7	+9.6	+9.6	+9.6
Rockies	7.2	+10.1	+10.8	+23.6
Southwest	2.5	+4.0	+3.6	+8.3
Northwest	0.6	+0.3	+0.3	-
Alaska	0.0	+1.0	+1.0	+1.0
Total, Rest of West	15.1	+30.0	+30.1	+47.2
Total U.S.	231.0	+124.0	+118.7	+120.2
B. COAL TRANSPORTATION[b/]				
Transportation Employment	14.5	+10.7	+10.8	+14.9
C. TOTAL, MINING AND TRANSPORTATION				
Coal Mining	231.0	+124.0	+118.7	+120.2
Transportation	14.5	+10.7	+10.8	+14.9
Total	245.5	+134.7	+129.5	+135.1

a/ Details in Attachment A-1.

b/ Details in Attachment A-2.

requirements without necessarily increasing total regional production. This is illustrated in regions such as Pennsylvania Central, where total production in 1995 is forecasted to decline from 1980 levels. However, in addition to this forecasted decline is a shift from higher-sulfur coals (which in general have higher productivities) to lower-sulfur coals (which in general have lower productivities). Because of these shifts, average productivity in these regions is forecast to decline from base case levels, resulting in a greater number of workers needed to support a lower level of production.

These types of intraregional shifts can be seen in Attachment A-3, where production and employment for an illustrative region (Indiana) are estimated by type of coal and mining method.

- In 1980, most of Indiana's production came from high-sulfur surface mines, with smaller amounts from underground and/or medium-sulfur mines. Stripping ratios ranged from between 12:1 and 25:1 (feet of overburden per foot of coal). Productivity at surface mines averaged about 5,800 tons per worker-year (or about 25 tons per worker-day), covering a range from less than 3,000 to more than 7,000 tons per worker-year.

- By 1995, some of these existing mines will have retired because of exhaustion of reserves. New replacement mines at a given sulfur level would generally have less favorable geologic characteristics, either surface mines which are smaller and/or have higher overburden ratios, or shifts to deep mines. In each case, the productivity of these replacement mines will tend to be less than that of the older, low-stripping ratio mines, thus requiring more workers for a given production level. For example, we forecast that by 1995 new, high-sulfur surface mines would have stripping ratios of about 25:1 and productivity of about 4,000 tons per worker-year (or about 19 tons per worker-day). At these stripping ratios, new deep mines are cost-competitive in seams about five to six feet thick, with a productivity of about 3,000 tons per worker-year (or about 14 tons per worker-day). These deep mines can be cost competitive despite the lower labor productivity because the surface mines are more capital and power intensive. Hence, a shift to deep production as surface reserves are depleted involves lower productivity and more miners, but without a substantial jump in price.

- In the cost-effective equivalent case, the lower production levels lead to fewer new mine openings, thereby slowing the decline in average productivity. However, the relative increase in demand for medium-sulfur coals observed in the cost-effective

equivalent case leads to a greater shift from high-sulfur to medium-sulfur coals. Such coals tend to be scarcer in states like Indiana, with thinner seams and/or higher stripping ratios. These mines would tend to have lower productivity than most of the high-sulfur mines, and hence require more employment for a given production level. For example, the medium-sulfur coals are forecasted to be from deep mines in seams about four feet thick. Productivity in these mines is forecasted to be about 2,000 tons per worker-year (or about 9 tons per worker-day), which is considerably lower than the 3,000 ton per worker-year productivities forecasted for high-sulfur deep mines in five to six feet thick seams.

This methodology for estimating coal mining employment represents an improvement over earlier methods which assumed constant productivity within a region over time. In particular, this methodology explicitly recognizes differences resulting from coal quality, method of mining, coal geology, levels of preparation, and mine retirements. This methodology is also sensitive to many key assumptions such as the extent and cost of surface-mineable coals and the rates of retirement for existing coal mines. Because information on such parameters is often sparse, employment forecasts must still be considered somewhat uncertain.

Some states include both higher-sulfur and lower-sulfur regions. Therefore, some of the production and employment shifts among regions would be intrastate. For example, shifts out of Kentucky West, a higher-sulfur region in the Midwest, would be offset in part by gains in Kentucky East, a lower-sulfur region in Central Appalachia. Shifts out of West Virginia North (in Northern Appalachia) would be offset in part by gains in West Virginia South (in Central Appalachia).

-- Coal Transportation

The production shifts forecasted under the cost-effective equivalent alternative would lead to greater requirements for coal transportation. As indicated in Attachment A-2, coal transportation requirements would rise by approximately 100 billion ton-miles in 1995 above the levels forecasted in both the base case and Waxman-Sikorski cases. This in turn would lead to about 4,000 additional jobs required for coal transportation, primarily rail and truck movements. Because of the estimating methodology for transportation employment used in this analysis focuses on those directly involved in the coal movements, we believe these estimates are conservative and likely to understate the number of additional workers needed.

-- Related Employment Changes

These estimated changes in coal mining and transportation employment do not include shifts in related industries. In those regions where mining employment losses occurred, there would likely be additional losses associated with the industries supplying those mines and workers. Conversely, there would likely be gains in industries supplying mines and workers in those regions which increased production. Similarly, changes in employment in related manufacturing industries (particularly those for scrubbers and mining equipment) were not assessed. Further, this analysis does not include any changes in employment and/or standards of living associated with the changes in total national costs of meeting coal and electricity demands.

ATTACHMENT A-1

ESTIMATED COAL PRODUCTION AND COAL MINING EMPLOYMENT
IN 1980 AND IN 1995 UNDER DIFFERENT SCENARIOS

Supply Region	Production (10^6 tons)				Average Number of Workers (10^3 workers)				Employment Change from 1980 to 1995 (10^3 workers)		
		1995 Forecast				1995 Forecast					
	1980	Base Case	Waxman-Sikorski	Cost-Effective Equivalent	1980	Base Case	Waxman-Sikorski	Cost-Effective Equivalent	Base Case	Waxman-Sikorski	Cost-Effect Equivalen
Northern Appalachia											
PC	61.4	56.8	51.6	50.2	22.1	26.7	23.6	23.7	4.5	1.4	1.5
IW	31.7	57.9	53.7	38.4	13.9	28.5	25.6	19.2	14.6	11.7	5.3
OH	39.4	41.2	45.5	19.4	14.7	15.0	16.4	6.9	0.3	1.7	-7.8
MD	3.8	2.9	2.9	2.3	1.1	1.2	1.2	0.9	0.1	0.1	-0.2
WN	48.8	54.2	48.4	46.5	17.3	22.2	19.4	19.5	4.9	2.1	2.2
TOTAL	185.1	213.0	202.2	156.8	69.1	93.7	86.1	70.2	24.5	17.0	1.1
Central Appalachia											
WS	72.7	135.4	140.4	163.5	36.9	70.0	73.2	84.1	33.1	36.3	47.2
VA	41.0	42.7	42.9	49.1	16.1	19.2	19.2	22.7	3.1	3.2	6.7
KE	109.2	106.8	107.6	126.6	35.7	40.4	40.7	49.5	4.7	5.0	13.8
TN	9.9	9.8	8.6	10.5	3.9	4.3	3.7	4.5	0.4	-0.2	0.6
TOTAL	232.8	294.8	299.4	349.8	92.6	133.9	136.9	160.8	41.3	44.3	68.2
Southern Appalachia											
AL	26.4	25.9	26.2	26.5	11.4	14.3	14.4	15.1	2.9	3.0	3.7
TOTAL	26.4	25.9	26.2	26.5	11.4	14.3	14.4	15.1	2.9	3.0	3.7
TOTAL APPALACHIA	444.3	533.6	527.8	533.2	173.1	241.8	237.4	246.2	68.7	64.3	73.1
Midwest											
IL	62.5	72.5	87.1	27.7	17.8	20.5	23.6	7.9	2.8	5.9	-9.8
IN	30.9	46.8	40.1	23.9	5.3	14.0	11.1	6.7	8.7	5.8	1.4
KW	41.0	62.6	59.7	36.6	12.0	17.6	16.5	10.4	5.5	4.5	-1.6
TOTAL	134.4	182.0	186.9	88.2	35.1	52.1	51.3	25.0	17.0	16.1	-10.1
TOTAL MIDWEST	134.4	182.0	186.9	88.2	35.1	52.1	51.3	25.0	17.0	16.1	-10.1
Eastern Northern Great Plains											
ND	17.0	32.7	32.6	32.7	1.7	3.0	3.0	3.0	1.3	1.2	1.3
ME	0.3	0.2	0.2	0.2	-	-	-	-	-	-	-
TOTAL	17.3	33.0	32.8	32.9	1.7	3.0	3.0	3.0	1.3	1.2	1.3
Western Northern Great Plains											
MW	-	1.7	1.7	1.7	-	0.4	0.4	0.4	0.4	0.4	0.4
MP	29.6	69.1	71.1	81.5	1.1	2.5	2.6	3.0	1.4	1.5	1.9
WP	68.8	129.8	127.7	129.5	2.0	3.8	3.7	3.8	1.8	1.7	1.8
WG	26.1	49.0	49.8	56.9	2.8	6.0	6.1	7.5	3.2	3.3	4.6
TOTAL	124.5	249.7	250.2	269.7	6.0	12.8	12.9	14.7	6.8	6.9	8.8
TOTAL NORTHERN GREAT PLAINS	141.7	282.6	283.1	302.5	7.7	15.8	15.8	17.7	8.1	8.1	10.0

ATTACHMENT A-1 (Continued)

ESTIMATED COAL PRODUCTION AND COAL MINING EMPLOYMENT
IN 1980 AND IN 1995 UNDER DIFFERENT SCENARIOS

Supply Region		Production (10^6 tons)				Average Number of Workers (10^3 workers)				Employment Change from 1980 to 19 (10^3 workers)		
			1995 Forecast				1995 Forecast					
		1980	Base Case	Waxman-Sikorski	Cost-Effective Equivalent	1980	Base Case	Waxman-Sikorski	Cost-Effective Equivalent	Base Case	Waxman-Sikorski	Cost-Effecti Equivalent
Central West												
	IA	0.6	1.0	1.0	1.0	0.1	0.3	0.3	0.3	0.2	0.2	0.2
	MO	5.5	6.9	6.9	4.4	1.1	1.8	1.8	1.0	0.7	0.7	-0.1
	KS	0.8	3.2	2.3	1.9	0.2	1.1	0.8	0.6	0.9	0.6	0.4
	AN	0.3	5.8	5.8	7.0	0.1	2.7	2.7	3.3	2.5	2.5	3.1
	OK	5.4	6.3	6.4	6.8	1.3	2.2	2.2	2.4	0.9	0.9	1.1
TOTAL		12.6	23.3	22.5	21.1	2.9	8.1	7.8	7.6	5.2	4.8	4.6
Gulf												
	TX	29.4	106.8	107.0	107.0	1.7	8.9	9.0	9.0	7.2	7.2	7.2
	LA	-	14.5	14.5	14.5	-	1.9	1.9	1.9	1.9	1.9	1.9
	AS	-	5.0	5.0	5.0	-	0.5	0.5	0.5	0.5	0.5	0.5
TOTAL		29.4	126.2	126.4	126.4	1.7	11.3	11.4	11.4	9.6	9.6	9.6
Rockies												
	CO	13.7	19.7	22.5	52.4	1.7	2.4	2.9	8.2	0.7	1.2	6.5
	ID	0.4	0.6	0.7	0.7	0.2	0.3	0.4	0.4	0.1	0.2	0.2
	MT	0.9	7.7	8.4	16.5	0.4	2.7	2.9	6.0	2.3	2.5	5.6
	UT	3.8	10.2	11.4	20.2	1.4	3.4	3.7	8.0	2.0	2.3	6.6
	CS	0.1	1.1	1.5	2.1	-	0.2	0.2	0.3	0.1	0.2	0.2
	UC	13.2	32.4	30.6	30.8	3.5	8.2	7.8	7.8	4.7	4.3	4.3
	US	-	0.8	2.9	2.1	-	-	0.3	0.2	-	0.3	0.2
TOTAL		32.1	72.5	78.1	132.7	7.2	17.3	18.0	30.8	10.1	10.8	23.6
Southwest												
	AZ	10.9	16.6	14.9	22.8	0.9	1.4	1.3	2.1	0.5	0.4	1.2
	NM	1.9	7.1	6.3	16.4	0.6	2.3	2.0	5.7	1.7	1.4	5.1
	NS	16.6	37.6	37.9	38.9	1.0	2.8	2.9	3.0	1.8	1.9	2.0
TOTAL		29.3	61.3	59.1	78.1	2.5	6.5	6.2	10.9	4.0	3.6	8.3
Northwest												
	WA	5.1	6.3	6.3	4.4	0.6	0.9	0.9	0.6	0.3	0.3	-
TOTAL		5.1	6.3	6.3	4.4	0.6	0.9	0.9	0.6	0.3	0.3	-
Alaska												
	AK	0.8	12.3	12.3	12.3	-	1.0	1.0	1.0	1.0	1.0	1.0
TOTAL		0.8	12.3	12.3	12.3	-	1.0	1.0	1.0	1.0	1.0	1.0
TOTAL WEST		109.3	301.9	304.8	375.1	15.1	45.1	45.2	62.2	30.0	30.1	47.2
TOTAL U.S.		829.7	1,300.1	1,302.5	1,299.0	231.0	354.8	349.7	351.2	124.0	118.7	120.2

Note: Totals may not add due to independent rounding.

ATTACHMENT A-2

ESTIMATED COAL TRANSPORTATION TON-MILES AND EMPLOYMENT
IN 1980 AND IN 1995 UNDER DIFFERENT SCENARIOS

	Ton-Miles ($\times 10^9$)				Average Number of Workers[a] (10^3 workers)				Employment Change from 1980 to 1995[a] (10^3 workers)		
		1995 Forecast				1995 Forecast					
	1980	Base Case	Waxman-Sikorski	Cost-Effective Equivalent	1980[b]	Base Case	Waxman-Sikorski	Cost-Effective Equivalent	Base Case	Waxman-Sikorski	Cost-Effective Equivalent
Mode											
Rail	N/A	462.2	461.4	557.6	6.1	11.6	11.6	13.4	5.5	5.5	7.3
Barge/Lake[c]	N/A	68.4	70.0	69.0	1.3	2.0	2.0	1.9	0.7	0.7	0.6
Truck	N/A	9.5	9.5	10.5	5.1	7.4	7.4	9.9	2.3	2.3	4.8
Other[d]	N/A	644.8	646.1	647.4	2.0	4.2	4.2	4.2	2.2	2.2	2.2
Total[e]		1,185.0	1,187.0	1,284.5	14.5	25.2	25.3	29.4	10.7	10.8	14.9

a/ Because the methodology focuses upon those workers directly involved in the movement of coal, total transportation employment requirements are likely to be underestimated.

b/ ICF estimates for 1980 employment were developed by applying forecasted transportation productivity (by origin-destination and mode of transport) to actual 1980 coal flows.

c/ Over many origin-destination routes, rail-barge and all-rail movements exist. In CEUM, these are generally represented as all-rail. As a result, forecasted barge movements are likely to be understated, and rail movements overstated by a similar amount.

d/ Mostly ocean transportation for exports.

e/ Totals may not add due to independent rounding.

ATTACHMENT A-3

PRODUCTION AND EMPLOYMENT SHIFTS IN INDIANA
IN 1980 AND IN 1995 UNDER DIFFERENT SCENARIOS

Sulfur Level	Production (10^3 tons)			Average Number of Workers			Productivity (tons/man-year)		
	Surface	Deep	Total	Surface	Deep	Total	Surface	Deep	Total
1980[a]									
Medium Sulfur	2,113	48	2,161	361	9	370	5,847	5,060	5,827
High-Medium Sulfur	2,415	55	2,470	413	11	424	5,847	5,060	5,827
High Sulfur	10,565	241	10,806	1,807	48	1,855	5,847	5,060	5,827
Very-High Sulfur	15,093	344	15,437	2,581	68	2,649	5,847	5,060	5,827
Total	30,185	688	30,873	5,162	136	5,298	5,847	5,060	5,827
Base Case -- 1995									
Medium Sulfur	2,097	3,443	5,540	626	1,431	2,056	3,350	2,407	2,694
High-Medium Sulfur	2,427	3,440	5,867	751	1,429	2,179	3,232	2,408	2,691
High Sulfur	14,972	3,760	18,732	4,364	1,331	5,695	3,431	2,824	3,289
Very-High Sulfur	11,638	5,025	16,663	2,519	1,516	4,035	4,620	3,315	4,130
Total	31,133	15,669	46,802	8,260	5,706	13,966	3,769	2,746	3,351
Waxman-Sikorski Case									
Medium Sulfur	1,444	3,079	4,524	367	1,262	1,630	3,931	2,439	2,776
High-Medium Sulfur	1,918	3,396	5,314	547	1,408	1,955	3,507	2,411	2,718
High Sulfur	5,818	71	5,890	1,295	12	1,307	4,494	5,827	4,506
Very-High Sulfur	19,392	5,025	24,417	4,676	1,516	6,191	4,148	3,315	3,944
Total	28,573	11,571	40,144	6,885	4,199	11,083	4,150	2,756	3,622
Cost-Effective Equivalent Case									
Medium Sulfur	2,589	3,443	6,033	823	1,430	2,254	3,145	2,407	2,677
High-Medium Sulfur	2,427	3,440	5,867	751	1,429	2,180	3,232	2,408	2,692
High Sulfur	3,478	71	3,549	597	12	609	5,827	5,827	5,827
Very-High Sulfur	8,374	101	8,475	1,634	17	1,651	5,126	5,827	5,134
Total	16,868	7,056	23,924	3,805	2,889	6,693	4,434	2,442	3,574

a/ 1980 values are estimates, reflecting incomplete data on production by sulfur level. Data were unavailable to vary average productivity in 1980 across sulfur levels.

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Written Statement for the Record
of
Clean Coal Coalition, Inc.

Before Joint Hearings of the
Subcommittee on Energy Development and Applications
and the
Subcommittee on Natural Resources, Agricultural Research
and Environment

on Acid Rain: Implications for R&D and Technology

October 4, 1983

The following written statement for the record is filed on behalf of the Clean Coal Coalition, Inc. (Coalition). This coalition is an organization of interested companies and organizations dedicated to the development of federal/state policies which encourage the use of more flexible, least-cost emissions control strategies under the Clean Air Act. Companies representing a variety of viewpoints drawn from the coal producer/fuel supplier, industrial user, electric utility, equipment manufacturing, gas utility, project development, engineering/construction and consulting communities comprise the Clean Coal Coalition, Inc.

Summary

Our written statement urges the Congress to avoid revisions to the Clean Air Act (Act) which would state reductions in sulfur dioxide emissions in terms of percentage reductions which would effectively mandate post-combustion cleanup and exclude more cost-beneficial, attractive alternatives. In this regard, the plant operator should be entitled under the Act to determine which control measures would be most effective and

sensible on the basis of site-specific characteristics of the plant. Flue gas scrubbing is one sulfur dioxide emission control option among several. Current federal statutory requirements should not operate to dictate flue gas scrubbing as the only emission control option, but should preserve appropriate flexibility for development and use of other less costly emission control strategies.

Finally, we would focus Congressional attention on the fact that conventional coal cleaning is and should be recognized as the most cost-effective first step toward control of sulfur dioxide emissions. Early commercial introduction of economical coal/water fuels using deeply cleaned coal can increase the effectiveness of conventional coal cleaning. This approach can be coupled with deployment of the select use of natural gas to augment cleaned coal as a cost-effective, secondary step toward control of sulfur dioxide emissions.

Introduction

To a coal-burning utility plant, the most undesirable impurity in its fuel heretofore has not been sulfur but ash-forming mineral matter which causes fouling and slagging of its boiler tubes. These phenomena degrade boiler performance due to reduced gas-to-tube heat transfer (hence increased heat rate to produce a given output), shorten intervals between de-slagging operations, and shorten tube life due to material attack (hence decreased boiler availability).

Cost of Reduced Coal Quality

> "The cheapest electricity is not of necessity produced from the cheapest coal" (1).

It has been observed repeatedly that the availability of U.S. generating stations has been deteriorating over the years with the exploitation of lower-grade coal resources, as

illustrated in Figure 1. As noted by a prominent electric utility executive, "The drop is very substantial having taken into account the effect of the increasing production of low Btu Powder River Basin coal" (1).

The cost to the utility of impaired plant availability is also substantial. In the case of the American Electric Power (AEP) system cited as an example, to make up a 10 percent loss in availability of its 1,455 MW(e) coal-fired generating capacity due to coal-related outages would equate to a cost in excess of $1 billion (1980) for the financing and construction of new facilities (1).

Conversely, a more specific case of benefit from improvement in coal quality by conventional washing has been cited for the Muskingum River station on the AEP system (2). In this case, an investment of $18 million in upgrading the preparation facility at its Central Ohio Coal Company permitted the capacity of one of its Ohio Power Company units to be upgraded from 535 to 585 MW(e). This increase in capacity, if new, would have required a capital expenditure of $55 million and an annual owning cost of $13.8 million. It also increased the plant availability by 5 percent, yielding a benefit in pre-tax income of approximately $6 million per year to the utility system (2).

Regional Coal Characteristics

The characteristics of U.S. coal vary widely according to region, bed, coal seam and location within a seam. The composite characteristics of raw coals by region collected and aggregated by the U.S. Bureau of Mines (3) are presented in Table 1. It may be noted that the energy content of the low-sulfur, predominantly sub-bituminous Western coals is somewhat lower than that of the Eastern bituminous coals even on a moisture-free basis. Because of the much greater moisture

content of the Western coals, however, their actual heating value is significantly lower than those of the Eastern coals.

The comparable characteristics of these coals following partial de-ashing in a routine preparation operation (crushing to 3/8-inch top size and heavy-media washing at 1.60 specific gravity) are presented in Table 2. It may be noted that, in addition to ash, the mineral (pyritic) sulfur content of the coals is reduced, thus together producing a greater difference in energy content between the Eastern and Western coals, even on a dry basis. Since the moisture content of the lower-rank Western coal is probably increased by washing somewhat more than that of the more highly developed Eastern coals, the spread in actual heating value would be increased still further. This penalty in moisture content and heating value led the Bureau of Mines investigators to conclude that "...therefore, considering also the high transportation costs, extensive use of these (Western) coals in Eastern utility plants is doubtful" (4).

Sulfur Emissions

So far nothing has been said about the potential of pre-combustion coal cleaning for reducing the emissions of sulfur dioxide released during combustion. Comparison of the data in Tables 1 and 2 suggest that it can be significant for those coals in which a high fraction of the sulfur content is pyritic in nature. This end result for the Northern Appalachian coals is particularly fortunate, since this is the region in which acid rain is most extensive, as shown in Figure 2, which is reproduced from the NAS/NRC report Acid Deposition (5).

The corresponding SO_2 emission levels in the same regions are shown in Figure 3. It may be seen that, in addition to the states in the northern Appalachian region (Ohio, Pennsylvania,

northern West Virginia), New York is also a significant SO_2 emitter. New York imports its coal, more than 80 percent of it from Pennsylvania with an average heat value of 12,045 Btu/lb and average sulfur and ash contents of 1.9 and 14.3 weight percent, respectively (6). Because of their high SO_2 emissions and close proximity upwind of the New England states most affected by acid rain, these four states constitute a regional group offering the best opportunity for a concentrated definitive experiment to determine the contribution of SO_2 emissions to acid precipitation.

Options for SO_2 Reduction

Four near-term options are immediately available as a means for effecting a sharp reduction in the level of SO_2 emissions in the four-state region identified above:

- Precombustion coal cleaning
- Select use of natural gas
- Switching to imported low-sulfur coal
- Flue gas desulfurization (scrubbing)

Of these options, the last two carry a cost penalty to the utilities involved (and their rate-payers). The scrubber option is the most costly. A study by the Pennsylvania Electric Company has shown that "on average the capital cost of an FGD system is $230/KW, more than ten times the cost of low- to mid-level PCC (physical coal cleaning) systems at $20/KW. An FGD system costs, on average, 10 mills/KWH annually to own and operate (first year costs) compared to 3.0 mills/KWH for a PCC system" (7). A third, unstated but large, cost of FGD systems in that they lower the overall availability of the powerplant. Contrary to the uninformed belief of some, scrubbers are not "mechanical devices...which filter out sulfur dioxide" (8) but complex chemical plants connected directly downstream of the powerplant itself. Their well-known

susceptibility to failure adversely affects overall system reliability and availability with the associated high cost penalty discussed earlier.

Switching to imported, low-sulfur coal might be lower in direct cost to the utility despite its already premium price and vulnerability to transportation rate increases by the railroads. However, the cost of breaking long-term contracts with local coal producers would surely be substantial. Still higher would be the unquantifiable social cost of disemploying local coal miners. The true cost of dislocating a family cannot be measured in terms of lost wages alone, as has been disingenuously proposed (8), with compensation paid to "retrain and help relocate" the unemployed.

Coal cleaning, on the other hand, is not a net cost but an investment leading to increased plant capacity and availability. If the AEP example cited earlier is any indicator, exercise of the coal-cleaning option can only improve the electric utility's "bottom line." Furthermore, it does not preclude additional control measures such as select use of natural gas for clean Btu augmentation or, as a last resort, scrubbing to the degree required.

Considering the above range of options, it is clear that to mandate one or another is neither desirable nor necessary. Given freedom of choice, the individual plant manager will determine the least-cost combination specific to his own requirements, and will start with coal cleaning. It is hardly surprising that the Acid Rain Peer Review Panel for the White House Office of Science and Technology Policy singled out "intensifying coal washing" as one of the "first, 'least-cost' steps" to reduce sulfur emissions (9, 10).

Limitations of Coal Cleaning

Why has the potential of physical coal cleaning (PCC) not previously received such high-level recognition? Physical methods for removing mineral impurities are effective on pyritic sulfur but do not affect the organic component, as may be concluded from comparing Tables 1 and 2. Depending upon the coal, accordingly, conventional cleaning can remove from 20 to 50 percent of the total sulfur (3) whereas a wet-sludge FGD system (when operating well) can remove up to 90 percent. This leads scrubber proponents to demand "Why not the best"? and the Clean Air Act to invoke superlative requirements for "Best Available Control Technology" (BACT) and "Lowest Achievable Emission Rate" (LAER) without explicitly calling for consideration of the costs involved.

Judged from a benefit/cost perspective by the Environmental Protection Agency, however, coal cleaning has in fact received high marks. But, they were reported to a limited audience. First considered as an adjunct to improve the effectiveness of FGD systems (11), PCC later received the ultimate accolade: "Physical coal cleaning is the least-cost method of reducing sulfur emissions from the combustion of coal" (12). This led the Coalition to coin the term "Least-Cost Compliance" for an investment strategy combining PCC with select use of natural gas (13).

Potential of Coal Cleaning

Studies for the Environmental Protection Agency indicated that conventional washing of all the utility coal delivered in 1979 from eight states comprising four Bureau of Mines regions could have achieved an additional reduction in SO_2 emissions of 2.8 million tons above that actually realized that year by cleaning only part of the coal (14). Subsequently, this appraisal was revised downward to 2.4 million tons (15), but a

more recent review adjusted the figure upward to about 2.5 million tons and extended the analysis to indicate that somewhat more intensive cleaning could double the number to 5 million tons annually (16).

These data are summarized in Table 3 together with the estimated cost of the two degrees of cleaning evaluated: conventional cleaning in which the coal is crushed to a top size of 1¼-inches and separated from the mineral matter in a heavy medium (a suspension of fine magnetite in water) with a specific gravity of 1.60, and a more intensive cleaning involving crushing to 3/8-inch top size and separating at a lower specific gravity of 1.30.

It may be seen that the major additional SO_2 reduction is achieved in the Northern Appalachian region whose coals are the most amenable to sulfur removal by physical means, as noted earlier. It may also be seen that the additional SO_2 reduction achievable by the more intensive cleaning is accompanied by a greatly increased cost of cleaning due to reduced "yield," or increased loss of energy value to the waste stream. This loss is illustrated graphically in Figure 4, which indicates that the decreased recovery (yield) is due not so much to the increased degree of comminution (size reduction) as to an excessively low specific gravity of separation. A more nearly optimum separation might appear to be at 1.40 specific gravity, yielding an 80 percent Btu recovery and a sulfur reduction of 41 percent. For comparison with the data of Table 3, such a degree of cleaning would produce an additional SO_2 reduction of about 3.5 million tons.

Elimination of Loss to Waste

A loss of 20 percent of coal energy value to the waste stream would be economically unacceptable to a commercial coal-cleaning operation. This loss can be eliminated, however,

if the waste is burned in an SO_2-capturing fluidized bed. Such an application could be designed to cogenerate high-temperature process heat for drying the washed coal, electric power to drive the mechanical operations at the preparation plant and for sale to the utility grid, and a benign solid combustion residue which need not be treated as a hazardous material but has useful commercial value (17). This subject is beyond the scope of the present discussion but has been elaborated elsewhere (18, 19).

Select Use of Natural Gas

The combined burning of coal with natural gas on a select use basis also appears to offer many potential economic, environmental and operational benefits to consumers, utilities, industrial users and the states. Under existing technology, coal can be used in a single combustor side-by-side with gas in another combustor, or coal and gas can be burned simultaneously in a single combustion. Further, sophisticated technological innovations may offer prospects for more effective long-term combined combustion applications (20).

With availability of technology and fuel supplies, the most significant impediment to broader deployment of select use is regulatory in nature. Regulatory approval of select use appears to be potentially achievable under both the Clean Air Act and the Powerplant and Industrial Fuel Use Act. But the regulatory review process is both formidable and uncertain, especially with respect to new facilities. (Figure 5) Specific uncertainty currently exists with respect to whether select use is recognized by EPA as a control technology on the basis of performance along with other technologies; whether a generic review process can be framed for such applications rather than a case-specific approach; and whether select use can be more effectively integrated into standards of BACT, RACT, LAER, and NSPS.

Costs and Potential for Application Appear Attractive

The number of states with high potential for deployment of select use of natural gas are fairly substantial and closely track the regional areas experiencing acid rain damage or symptoms. (Figure 6) The costs of sulfur dioxide control using select use may vary from $50/ton of sulfur dioxide removed to over $1,000/ton. By comparison, several other alternative control strategies range from $250-$2,000/ton indicating that select use like coal washing comprise the least cost approaches in the wide spectrum of alternatives (21). 1/

Conclusion

The Coalition maintains that low cost coal washing can achieve sulfur dioxide reductions of 2.5 million tons/year while deeper coal cleaning techniques could elevate such reductions to almost 3.5 million tons/year. The potential for select use could range from 1.5 million tons to 2.5 million tons/year depending upon the scope and breadth of application.

Therefore, the Coalition respectfully suggests that fuller scale deployment of these currently available, cost-effective control techniques should not be effectively limited by the types of legislation currently pending before the Congress (H.R. 3251, H.R. 3400; S. 145, S. 769).

1/ The Coalition strongly doubts that massive switching to low sulfur coals or even fuel blending would achieve levels of sulfur dioxide reductions without significant increases in the delivered costs of low sulfur coal (minemouth plus rail charges). Moreover, in several instances, this market subject to captive coal rail carriers could exceed 90% with such a development exerting a further gridlock on escalating rail charges for coal shipments. Further, there are other costs associated with heavy switching to low sulfur coal which have not been meaningfully considered in various analyses conducted to date including costs of displacement of eastern miners, plant derating, lower Btu content of fuels, and increases in solid waste disposal.

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TABLE 1

COMPOSITE RAW COAL CHARACTERISTICS
(Bureau of Mines Report of Investigation 8118, 1976)

Region	Moisture (Percent)	Ash (Percent)	Sulfur (Percent) Pyritic	Sulfur (Percent) Total	Btu per Pound (10^3)*
Northern Appalachian	1.7	15.1	2.01	3.01	12.7
Southern Appalachian	1.8	11.0	0.37	1.04	13.3
Alabama	1.4	9.5	0.69	1.33	13.7
Eastern Midwest	4.9	14.2	2.29	3.92	12.2
Western Midwest	4.0	16.2	3.58	5.25	12.1
Western	8.5**	8.9	0.23	0.68	12.4

*Moisture-free basis

**May increase significantly during transportation to user

TABLE 2

COMPOSITE WASHED COAL CHARACTERISTICS
(Crushed to 3/8-inch top size, cleaned at 1.60 specific gravity)

Region	Moisture (Percent)	Ash (Percent)	Sulfur (Percent) Pyritic	Sulfur (Percent) Total	Btu per Pound (10^3)*
Northern Appalachian	Increased	8.0	0.85	1.86	13.8
Southern Appalachian	Increased	5.1	0.19	0.91	13.2
Alabama	Increased	5.8	0.49	1.16	14.3
Eastern Midwest	Increased	7.5	1.03	2.73	13.1
Western Midwest	Increased	8.3	1.80	3.59	13.2
Western	Increased	6.3	0.10	0.56	12.8

*Moisture-free basis

TABLE 3

ADDITIONAL SO_2 REDUCTION ACHIEVABLE IN 1979 BY CLEANING ALL CLEANABLE UTILITY COAL IN FOUR REGIONS

Coal Source (Bu Mines Region)	Reduction By Conventional Cleaning (10^3 tons) [1/]	Average Annualized Cost of Cleaning ($/Clean Ton) [3/]	Reduction by More Intensive Cleaning (10^3 ton) [2/]	Approx. Increase Over Conventional ($/Clean Ton)
Northern Appalachia	1,420	7.53	3,360	12.27
Eastern Midwest	910	5.10	1,460	8.47
Southern Appalachia	150	7.63	530	9.63
Alabama	65	6.70	160	11.00
Total or Average	2,545	6.74	5,010	10.34

1/ Conventional Cleaning: Crush to 1½" top size; separate at 1.60 specific gravity.

2/ More Intensive Cleaning: Crush to 3/8" top size; separate at 1.30 specific gravity.

3/ Analytical Approximation:

$$\text{Cost of Cleaning} = \text{Operational Cost} + \frac{\text{Spot Price of Coal}}{\text{Yield}} (1 - \text{Yield})$$

Note: Cost of waste disposal not included.

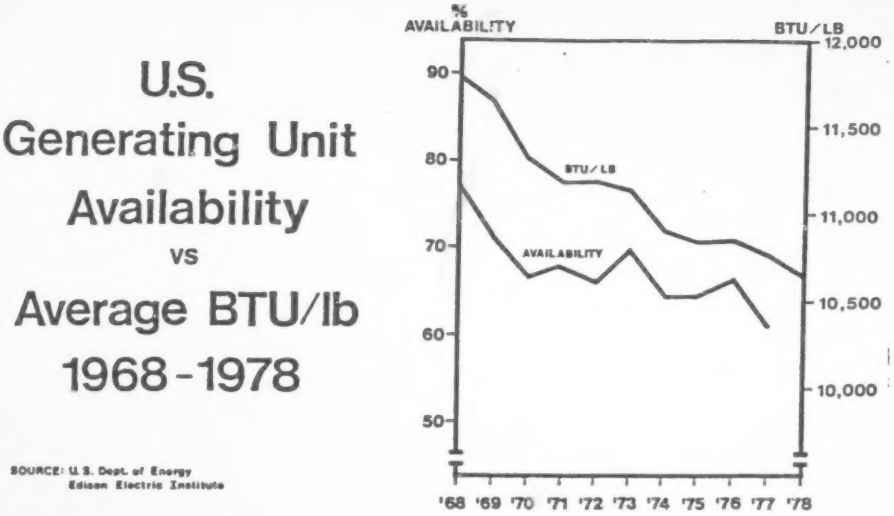

Figure 1. Decline in steam coal quality and availability of U.S. power plants (from Blackmore)

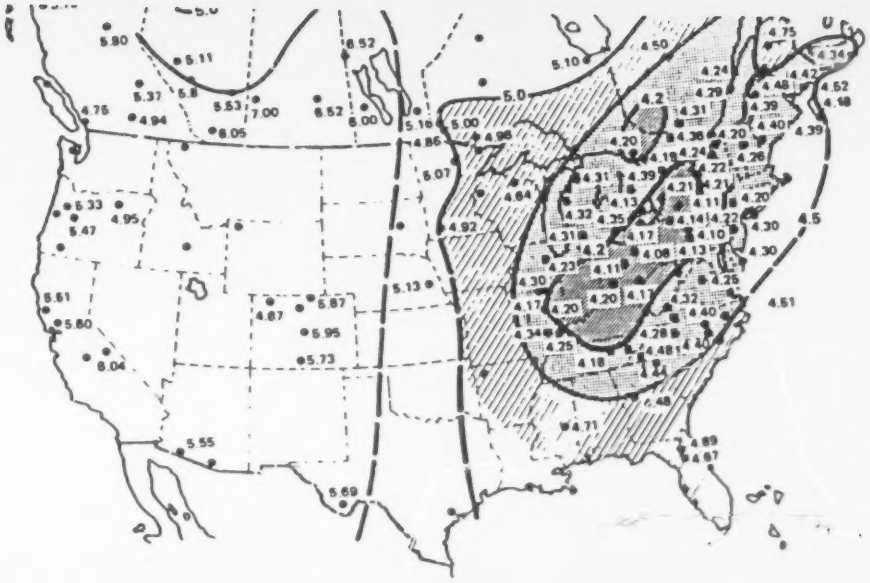

Figure 2. Annual mean pH in precipitation in U.S. and Canada for 1980.

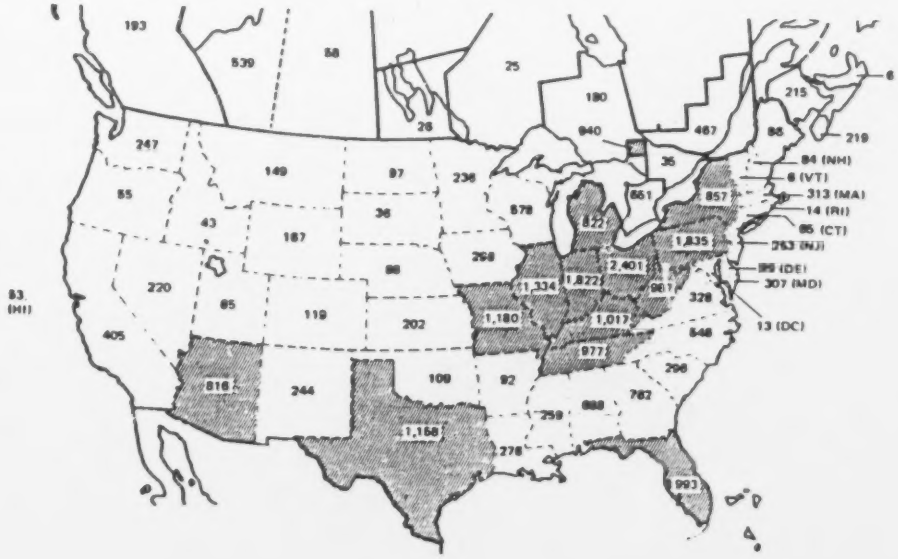

Figure 3. Representative values of SO_2 emissions in U.S. and Canada in 1980 (thousands of tonnes)

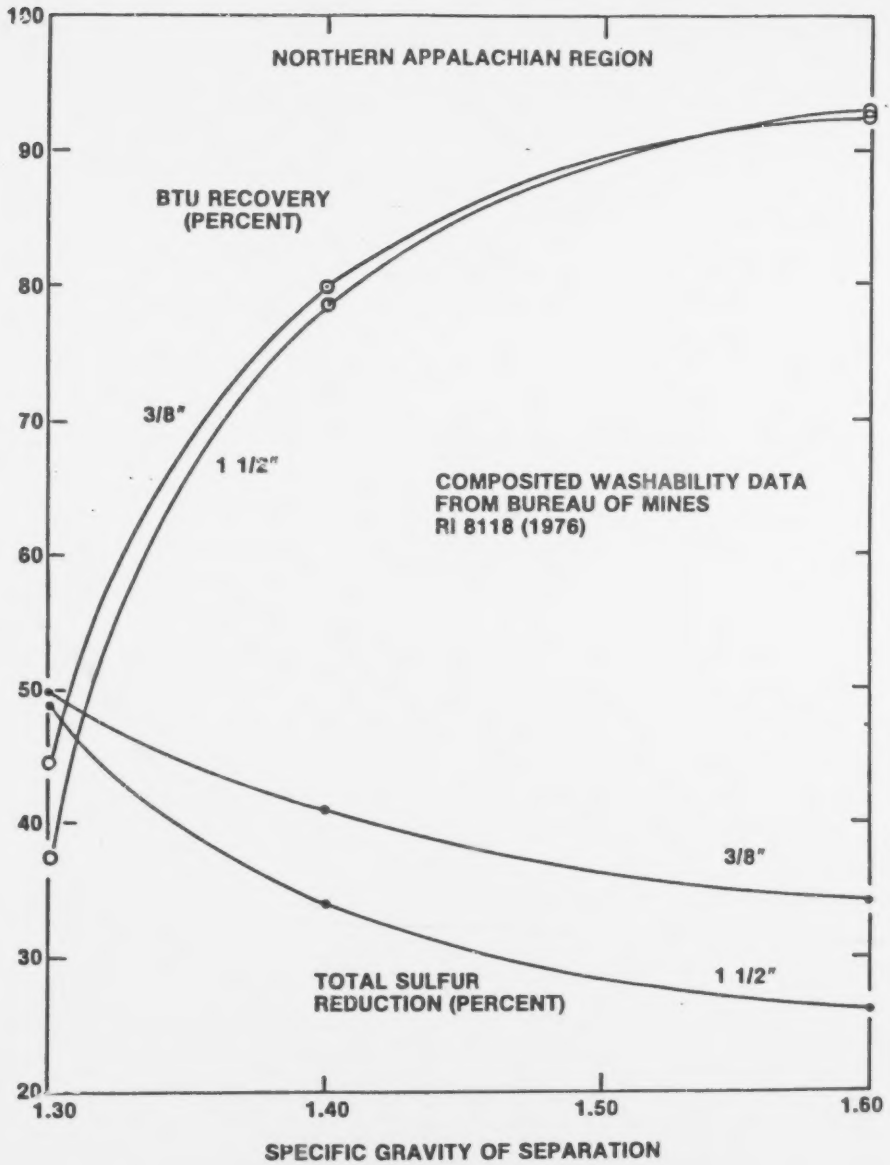

Figure 4. Sulfur reduction potential of Northern Appalachian coals subject to conventional washing.

REGULATORY IMPACTS ON THE POTENTIAL FOR SELECT GAS USE

Market	Impact of Clean Air Act	Impact of Fuel Use Act	Combined Impact
Existing Electric Powerplant	●	●	●
Existing Large Boiler	●	●	●
Existing Small Boiler	●	●	●
Existing Large Non-Boiler	●	●	●
Existing Small Non-Boiler	●	●	●
New Small Boiler	◒	●	◒
New Small Non-Boiler	△	●	△
New Large Non-Boiler	▲	●	▲
New Large Boiler	▲	△	▲
New Electric Powerplant	▲	△	▲

Legend

● Not a Constraint
◒ Moderate Constraint, Sizable Market
△ Moderate Constraint, Limited Market
▲ Severe Constraint

Note: Judgment Based on Current Interpretation of Legislation

(Figure 5)

STATES WITH HIGH POTENTIAL FOR SELECT GAS USE

Note: States shaded have relatively strict sulfur emission limits (oil <1 #/MMBtu; coal < 1.2 #/MMBtu) and also have either a heavy dependence on oil for electrical generation (> 10%), or have powerplants which are not currently meeting their sulfur emission limit.

(Figure 6)